

JP 3998903 B2 2007.10.31

(19) 日本特許庁(JP)

(12) 特 許 公 報 (B2)

(11) 特許番号

特許第3998903号
(P3998903)

(45) 発行日 平成19年10月31日(2007.10.31)

(24) 登録日 平成18年8月17日(2007.8.17)

(51) Int. Cl.

F J

C07C 211/61 (2006.01)
C09K 11/06 (2006.01)
H01L 51/30 (2006.01)C07C 211/61
C09K 11/06 620
C09K 11/06 660
H05B 33/14 B
H05B 33/22 D

請求項の数 6 (全 15 項)

(21) 出願番号 特願2000-268833 (P2000-368833)
(22) 出願日 平成12年9月5日(2000.9.5)
(65) 公開番号 特開2002-80433 (P2002-80433A)
(43) 公開日 平成14年3月19日(2002.3.19)
審査請求日 平成16年1月13日(2004.1.13)

前置審査

(73) 特許権者 000183646
出光興産株式会社
東京都千代田区丸の内3丁目1番1号
(74) 代理人 100078732
弁理士 大谷 保
(72) 発明者 植川 雄彌
千葉県旭ヶ浦市上泉1280番地
(72) 発明者 舟橋 正和
千葉県旭ヶ浦市上泉1280番地
審査官 天野 繁樹

最終頁に続く

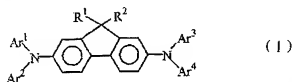
(54) 【発明の名称】 新規アリールアミン化合物及び有機エレクトロルミネッセンス素子

(57) 【特許請求の範囲】

【請求項1】

下記一般式(1)で表される新規アリールアミン化合物。

【化1】



19

(式中、 R^1 及び R^2 は、それぞれ独立に、置換もしくは未置換の炭素原子数1〜30のアルキル基、置換もしくは未置換の炭素原子数1〜30のアルコキシ基、置換もしくは未置換の炭素原子数6〜40のアリール基、置換もしくは未置換の炭素原子数7〜40のアリールアルキル基又は置換もしくは未置換の炭素原子数6〜40のアリールオキシ基を表す。

$Ar^1 \sim Ar^4$ は、それぞれ独立に、置換もしくは未置換の炭素原子数6〜40のアリール基を表し、それぞれ同一でも異なってもよい。ただし、 $Ar^1 \sim Ar^4$ のうち少なくとも2つは、置換もしくは未置換のm-ピフェニルで、残りは置換もしくは未置換の

20

(2)

JP 3998903 B2 2007.10.31

ビフェニルである。)

【請求項2】

前記一般式(1)の新規アリアルアミン化合物において、 $A\text{r}^1$ 及び $A\text{r}^2$ が置換もしくは未置換のm-ビフェニル、 $A\text{r}^3$ 及び $A\text{r}^4$ が置換もしくは未置換のビフェニルであることを特徴とする請求項1に記載の新規アリアルアミン化合物。

【請求項3】

請求項1に記載の一般式(1)で表される新規アリアルアミン化合物からなる有機エレクトロルミネッセンス素子用材料。

【請求項4】

一対の電極間に有機化合物層を有する有機エレクトロルミネッセンス素子であって、該有機化合物層が請求項3に記載の有機エレクトロルミネッセンス素子用材料を含有することを特徴とする有機エレクトロルミネッセンス素子。

【請求項5】

前記有機化合物層が、発光層又は正孔輸送層であることを特徴とする請求項4に記載の有機エレクトロルミネッセンス素子。

【請求項6】

一対の電極間に有機化合物層を有する有機エレクトロルミネッセンス素子であって、該有機化合物層が請求項3に記載の有機エレクトロルミネッセンス素子用材料と発光材料とを含有する層を有することを特徴とする有機エレクトロルミネッセンス素子。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】

本発明は新規アリアルアミン化合物及び有機エレクトロルミネッセンス素子に関し、特に、高輝度で、耐熱性が高く、長寿命で、正孔輸送性が優れ高発光効率な有機エレクトロルミネッセンス素子及びそれを実現する新規アリアルアミン化合物に関するものである。

【0002】

【従来の技術】

有機物質を使用した有機エレクトロルミネッセンス(EL)素子は、壁紙テレビの平面発光体やディスプレイのバックライト等の光源として使用され、盛んに開発が行われている。

有機材料の電界発光現象は、1963年にポーブ(Pope)らによってアントラセン単結晶で観測され(J. Chem. Phys. 38(1963)2042)、1965年にヘルフリッヒ(Helfrich)とシュナイダー(Schneider)は注入効率の良い溶液電極系を用いることにより比較的に強い注入型ELの観測に成功している(Phys. Rev. Lett. 14(1965)229)。それ以来報告されている様に、共役の有機ホスト物質と縮合ベンゼン環を持つ共役の有機活性化合物とで有機発光性物質を形成した研究が行われ、ナフタレン、アントラセン、フェナントレン、テトラセン、ピレン、ベンゾピレン、クリセン、ピセン、カルバゾール、フルオレン、ビフェニル、ターフェニル、トリフェニレンオキサライド、ジハロビフェニル、トランススチルベン及び1,4-ジフェニルブタジエン等が有機ホスト物質の例として示され、アントラセン、テトラセン及びペンタセン等が活性化合物の例として挙げられた。しかしこれらの有機発光性物質はいずれも $1\mu\text{m}$ を越える厚さを持つ単一層として存在し、発光には高電圧が必要であった。このため、真空蒸着法による薄膜素子の研究が進められた(例えばThin Solid Films 94(1982)171)。しかし、薄膜化は駆動電圧の低減には有効であったが、実用レベルの高輝度の素子を得るには至らなかった。

そこでタン(Tang)らは、陽極と陰極との間に2つの極めて薄い膜(正孔輸送層と発光層)を真空蒸着で積層したEL素子を考案し、低い駆動電圧で高輝度を実現した(Appl. Phys. Lett. 51(1987)913もしくは米国特許4356429号)。その後、正孔輸送層と発光層に用いる有機化合物の開発が十数年間進められた結果、実用化レベルの寿命と発光効率が達成された。その結果、有機EL素子は、カーステレ

(3)

JP 3998903 B2 2007.10.31

す、携帯電話の表示部などから実用化が開始されている。

【0003】

しかしながら、実用面において、発光輝度、長時間使用に対する経時劣化の耐久性などが十分ではなく、さらなる向上が求められている。特に、フルカラーディスプレイ等への応用を考えた場合には、R、G、Bの各色に対して、 300 cd/m^2 以上の高輝度で数千時間以上の半減寿命を到達することが求められている。これを実現するのが特に困難なのは、青色発光であり、青色発光させるには発光層のエネルギーギャップが 2.8 eV 以上と大きく、正孔輸送層と発光層の間にある正孔注入の際のエネルギー障壁が大きいので、界面に印加される電界強度は大きく、従来の正孔輸送層では安定に正孔注入ができず改良が求められていた。

また、有機EL素子を草搭載することを前提とした場合、 100°C 以上の高温保存性能に問題があることが指摘されている。この際も従来の正孔輸送層においてはガラス転移温度が低いことが指摘されており、これを 100°C 以上に改良することのみで対応しようとしたが、不十分であり高温における良好な保存性能は未だ実現していなかった。さらに、正孔輸送層と発光層との相互作用としてエキサイプレックスが生成して、素子の輝度が劣化するという問題もあった。

【0004】

【発明が解決しようとする課題】

本発明は、前記の課題を解決するためになされたもので、高輝度で、耐熱性が高く、長寿命で、正孔輸送性が優れ高発光効率な有機エレクトロルミネッセンス素子及びそれを実現する新規アリアルアミン化合物を提供することを目的とするものである。

【0005】

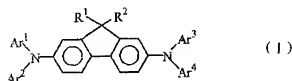
【課題を解決するための手段】

本発明者らは、前記の好ましい性質を有する有機エレクトロルミネッセンス素子（以下、有機EL素子）を開発すべく鋭意研究を重ねた結果、特定の構造を有する新規アリアルアミン化合物を有機化合物層に添加すると有機エレクトロルミネッセンス素子の輝度、耐熱性、寿命が向上し、さらに正孔輸送性が向上して高発光効率となることを見出し本発明を完成するに至った。

【0006】

すなわち、本発明は、下記一般式（1）で表される新規アリアルアミン化合物を提供するものである。

【化2】



【0007】

式中、 R^1 及び R^2 は、それぞれ独立に、置換もしくは未置換の炭素原子数1～30のアルキル基、置換もしくは未置換の炭素原子数1～30のアルコキシ基、置換もしくは未置換の炭素原子数6～40のアリアル基、置換もしくは未置換の炭素原子数7～40のアリアルアルキル基又は置換もしくは未置換の炭素原子数6～40のアリアルオキシ基を表す。

$Ar^1 \sim Ar^4$ は、それぞれ独立に、置換もしくは未置換の炭素原子数6～40のアリアル基を表し、それぞれ同一でも異なってもよい。ただし、 $Ar^1 \sim Ar^4$ のうち少なくとも2つは、置換もしくは未置換のm-ピフェニルで、残りは置換もしくは未置換のピフェニルである。

(4)

JP 3968903 B2 2007.10.31

また、本発明は、一般式(1)で表される新規アリールアミン化合物からなる有機EL素子用材料、並びに、一対の電極間に有機化合物層を有する有機EL素子であって、該有機化合物層が前記有機EL素子用材料を含有することを特徴とする有機EL素子をも提供するものである。

【0008】

【発明の実施の形態】

本発明の新規アリールアミン化合物は、上記一般式(1)で表される。

一般式(1)において、 R^1 及び R^2 は、それぞれ独立に、置換もしくは未置換の炭素原子数1～30のアルキル基、置換もしくは未置換の炭素原子数1～30のアルコキシ基、置換もしくは未置換の炭素原子数6～40のアリール基、置換もしくは未置換の炭素原子数7～40のアリールアルキル基又は置換もしくは未置換の炭素原子数6～40のアリールオキシ基を表す。

アルキル基としては、例えばメチル、エチル、*n*-プロピル、*i*so-プロピル等、アルコキシ基としては、メトキシ、エトキシ等、アリール基としてはフェニル、ビフェニル、ナフチル等、アリールアルキル基としては、ベンジル、 α -メチルベンジル、 α -エチルベンジル、 α 、 α -ジメチルベンジル、4-メチルベンジル、4-エチルベンジル、2-tert-ブチルベンジル、4-*n*-オクチルベンジル、ナフチルメチル、ジフェニルメチル等、アリールオキシ基としては、フェノキシ、ナフチルオキシ、アンスルオキシ、ピレニルオキシ、ビフェニルオキシ、フルオランテニルオキシ、クリセニルオキシ、バリレニルオキシ等が挙げられる。

【0009】

また、これら各基の置換基としては、例えばフッ素原子、塩素原子、臭素原子、沃素原子等のハロゲン原子、メチル、エチル、*n*-プロピル、*i*so-プロピル等のアルキル基、メトキシ、エトキシ等のアルコキシ基、フェノキシ等のアリールオキシ基、ベンジル、フェニル、フェニルプロピル等のアリールアルキル基、ニトロ基、シアノ基、ジメチルアミノ基、ジベンジルアミノ基、ジフェニルアミノ基、モルホリノ基等の置換アミノ基、フェニル基、トリル基、ビフェニル基、ナフチル基、アンスル基、ピレニル基等のアリール基、ビリジル基、チエニル基、フリル基、キノリル基、カルバゾリル基等のヘテロ環基などが挙げられる。

【0010】

また、一般式(1)において、 $A_{R^1} \sim A_{R^4}$ は、それぞれ独立に、置換もしくは未置換の炭素原子数6～40のアリール基を表し、それぞれ同一でも異なってもよい。

アリール基としては、例えばフェニル基、トリル基、ビフェニル基、ナフチル基、アンスル基、ピレニル基等が挙げられる。

一般式(1)において、 $A_{R^1} \sim A_{R^4}$ のうち少なくとも2つは、置換もしくは未置換の*m*-ビフェニルで、残りは置換もしくは未置換のビフェニルである。

【0011】

$A_{R^1} \sim A_{R^4}$ を表す基における置換基としては、例えばフッ素原子、塩素原子、臭素原子、沃素原子等のハロゲン原子、メチル、エチル、*n*-プロピル、*i*so-プロピル等のアルキル基、メトキシ、エトキシ等のアルコキシ基、フェノキシ等のアリールオキシ基、ベンジル、フェニル、フェニルプロピル等のアリールアルキル基、ニトロ基、シアノ基、ジメチルアミノ基、ジベンジルアミノ基、ジフェニルアミノ基、モルホリノ基等の置換アミノ基、フェニル基、トリル基、ビフェニル基、ナフチル基、アンスル基、ピレニル基、フルオランテニル基等のアリール基、ビリジル基、チエニル基、フリル基、キノリル基、カルバゾリル基等のヘテロ環基などが挙げられる。

さらに、アリール置換ビフェニルにおけるアリール置換基としては、例えばフェニル基、ビフェニル基、ターフェニル基、ナフチル基、アンスル基、フルオレニル基等が挙げられる。

A_{R^1} 及び A_{R^3} は、置換もしくは未置換の*m*-ビフェニル、 A_{R^2} 及び A_{R^4} は、置換もしくは未置換のビフェニルであることが好ましい。

(5)

JP 3998903 B2 2007.10.31

【0012】

本発明の有機EL素子は、一対の電極間に有機化合物層を有する有機EL素子であって、該有機化合物層が前記新規アリールアミン化合物の有機EL素子用材料を含有する。前記有機化合物層が発光層又は正孔輸送層であることが好ましい。また、前記有機化合物層が前記新規アリールアミン化合物の有機EL素子用材料と発光材料とを含有する層を有することも好ましい。

前記新規アリールアミン化合物を、有機化合物層の少なくとも一層に含有させると有機EL素子の輝度、耐熱性、寿命、発光効率が向上するのは、該アリールアミン化合物が、正孔輸送性に優れ、安定に正孔注入ができる上、ガラス転移点が高く、発光材料と相互作用しにくく、相互作用により生じる無輻射遷位が避けられるからである。

19

【0013】

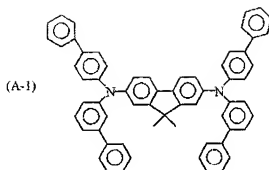
以下に、本発明の新規アリールアミン化合物において、一般式(1)の代表例を例示するが、本発明はこれらの代表例に限定されるものではない。

【0014】

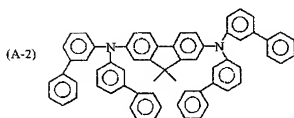
【化3】

(6)

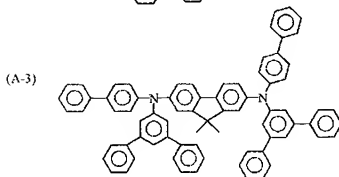
JP 3998903 B2 2007.10.31



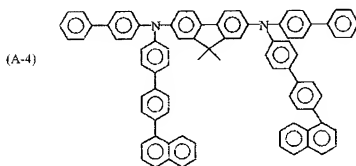
10



20



30

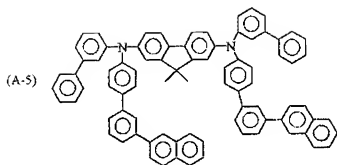


40

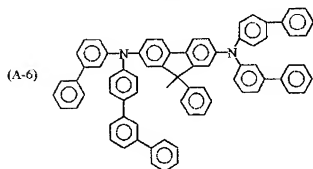
[0 0 1 5]
[1 4]

(7)

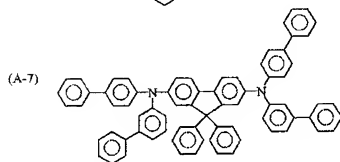
JP 3998903 B2 2007.10.31



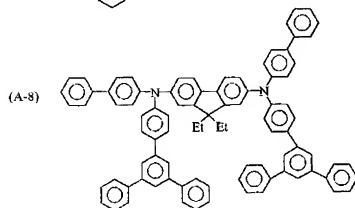
10



20



30

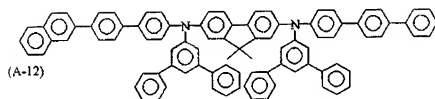
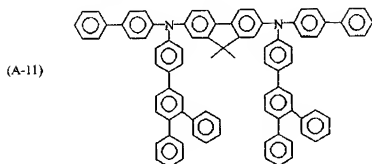
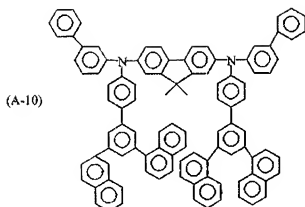


40

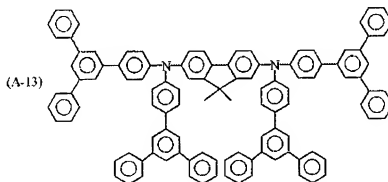
[0016]
[化5]

(8)

JP 3998903 B2 2007.10.31



[0 0 1 7]
[1 1 6]



50

(9)

JP 3998903 B2 2007.10.31

【0018】

本発明の有機E1素子は、陽極と陰極間に単層もしくは多層の有機化合物層を形成した素子である。単層型の場合、陽極と陰極との間に発光層を設けている。発光層は、発光材料を含有し、それに加えて陽極から注入した正孔、もしくは陰極から注入した電子を発光材料まで輸送させるために、正孔注入材料もしくは電子注入材料を含有しても良い。しかしながら、発光材料は、極めて高い蛍光量子効率、高い正孔輸送能力および電子輸送能力を併せ持ち、均一な薄膜を形成することが好ましい。多層型の場合E1素子は、(陽極/正孔注入層/発光層/陰極)、(陽極/発光層/電子注入層/陰極)、(陽極/正孔注入層/発光層/電子注入層/陰極)の多層構成で積層したものがある。

【0019】

発光層には、必要に応じて、本発明の新規アリールアミン化合物に加えてさらなる公知の発光材料、ドーピング材料、正孔注入材料や電子注入材料を使用することもできる。この新規アリールアミン化合物の好ましい使用法としては、発光層、電子注入層、正孔輸送層又は正孔注入層のいずれかの層に、濃度0.5～100重量%で添加する。さらに好ましくは、濃度50～100重量%である。

有機E1素子は、多層構造にすることにより、クエンチングによる頻度や寿命の低下を防ぐことができる。必要があれば、発光材料、他のドーピング材料、正孔注入材料や電子注入材料を組み合わせて使用することができる。また、他のドーピング材料により、発光頻度や発光効率の向上、赤色や白色の発光を得ることもできる。また、正孔注入層、発光層、電子注入層は、それぞれ二層以上の層構成により形成されても良い。その際には、正孔注入層の場合、電極から正孔を注入する層を正孔注入層、正孔注入層から正孔を受け取り発光層まで正孔を輸送する層を正孔輸送層と呼ぶ。同様に、電子注入層の場合、電極から電子を注入する層を電子注入層、電子注入層から電子を受け取り発光層まで電子を輸送する層を電子輸送層と呼ぶ。これらの各層は、材料のエネルギー準位、耐熱性、有機化合物層もしくは金属電極との密着性等の各要因により選択されて使用される。

【0020】

下記新規アリールアミン化合物と共に有機化合物層に使用できる発光材料またはホスト材料としては、縮合多環芳族族があり、例えばアントラセン、ナフタレン、フェナントレン、ピレン、テトラセン、ペンタセン、コロネン、クリセン、フルオレセイン、ペリレン、ルブレン及びそれらの誘導体がある。さらに、フタロペリレン、ナフトロペリレン、ペリノン、フタロペリノン、ナフトロペリノン、ジフェニルブタジエン、テトラフェニルブタジエン、クマリン、オキサジアゾール、アルダジン、ビスベンゾキサゾリン、ビスチリル、ピラジン、シクロペンタジエン、キノリン金属錯体、アミノキノリン金属錯体、ベンゾキノリン金属錯体、イミン、ジフェニルエチレン、ビニルアントラセン、ジアミノカルバゾール、ピラン、チオピラン、ポリメチン、メロシアン、イミダゾールキレート化オキシノイド化合物、キナクリドン、ルブレン、スチルベン系誘導体及び蛍光色素等が挙げられるが、これらに限定されるものではない。

【0021】

公知の正孔注入材料としては、正孔を輸送する能力を持ち、陽極からの正孔注入効果、発光層または発光材料に対して優れた正孔注入効果を有し、発光層で生成した励起子の電子注入層または電子注入材料への移動を防止し、かつ薄膜形成能力の優れた化合物が好ましい。具体的には、フタロシアニン誘導体、ナフトロシアニン誘導体、ポルフィリン誘導体、オキサゾール、オキサジアゾール、トリアゾール、イミダゾール、イミダゾロン、イミダゾールチオン、ピラゾリン、ピラゾロン、テトラヒドロイミダゾール、オキサゾール、オキサジアゾール、ヒドラゾン、アシルヒドラゾン、ポリアリールアルカン、スチルベン、ブタジエン、ベンジジン型トリフェニルアミン、スチルアミン型トリフェニルアミン、ジアミン型トリフェニルアミン等と、それらの誘導体、およびポリビニルカルバゾール、ポリシラン、導電性高分子等の高分子材料が挙げられるが、これらに限定されるものではない。

【0022】

(10)

JP 3998903 B2 2007.10.31

本発明の有機EL素子において使用できる公知の正孔注入材料の中で、さらに効果的な正孔注入材料は、芳香族三級アミン誘導体もしくはフタロシアニン誘導体である。

芳香族三級アミン誘導体の具体例は、トリフェニルアミン、トリトリルアミン、トリルジフェニルアミン、 $N, N', -ジフェニル-N, N' - (3-メチルフェニル) - 1, 1' - \text{ビフェニル} - 4, 4' - \text{ジアミン}$, $N, N, N', N' - (4-メチルフェニル) - 1, 1' - \text{フェニル} - 4, 4' - \text{ジアミン}$, $N, N, N', N' - (4-メチルフェニル) - 1, 1' - \text{ビフェニル} - 4, 4' - \text{ジアミン}$, $N, N' - \text{ジフェニル}-N, N' - \text{ジナフチル} - 1, 1' - \text{ビフェニル} - 4, 4' - \text{ジアミン}$, $N, N' - (メチルフェニル) - N, N' - (4-n-ブチルフェニル) - \text{フェナントレン} - 9, 10 - \text{ジアミン}$, $N, N - \text{ビス} (4-ジ-4-トリルアミノフェニル) - 4 - \text{フェニル}-シクロヘキサン等、もしくはこれらの芳香族三級アミン骨格を有したオリゴマーもしくはポリマーであるが、これらに限定されるものではない。$

フタロシアニン (Pc) 誘導体の具体例は、 $H_2 Pc$, $CuPc$, $CoPc$, $NiPc$, $ZnPc$, $PdPc$, $FePc$, $MnPc$, $ClAlPc$, $ClGaPc$, $ClInPc$, $ClSnPc$, $ClSiPc$, $(HO) AlPc$, $(HO) GaPc$, $VOpc$, $TiOPc$, $MoOPc$, $GaPc-O-GaPc$ 等のフタロシアニン誘導体およびナフタロシアニン誘導体であるが、これらに限定されるものではない。

[0023]

公知の電子注入材料としては、電子を輸送する能力を持ち、極陰からの電子注入効果、発光層または発光材料に対して優れた電子注入効果を有し、発光層で生成した励起子の正孔注入層への移動を防止し、かつ薄膜形成能力の優れた化合物が好ましい。具体的には、フルオレノン、アントラキノジメタン、ジフェノキノン、チオピランジオキシド、オキサゾール、オキサジアゾール、トリアゾール、イミダゾール、ペリレンテトラカルボン酸、フレオレニリデンメタン、アントラキノジメタン、アントロン等とそれらの誘導体が挙げられるが、これらに限定されるものではない。また、正孔注入材料に電子受容物質を、電子注入材料に電子供与性物質を添加することにより電荷注入性を向上させることもできる。

[0024]

本発明の有機EL素子において、さらに効果的な公知の電子注入材料は、金属錯体化合物もしくは含窒素五員環誘導体である。

金属錯体化合物の具体例は、8-ヒドロキシキノリナートリチウム、ビス (8-ヒドロキシキノリナート) 亜鉛、ビス (8-ヒドロキシキノリナート) 銅、ビス (8-ヒドロキシキノリナート) マンガン、トリス (8-ヒドロキシキノリナート) アルミニウム、トリス (2-メチル-8-ヒドロキシキノリナート) アルミニウム、トリス (8-ヒドロキシキノリナート) ガリウム、ビス (10-ヒドロキシベンゾ [h] キノリナート) ペリリウム、ビス (10-ヒドロキシベンゾ [h] キノリナート) 亜鉛、ビス (2-メチル-8-キノリナート) クロロガリウム、ビス (2-メチル-8-キノリナート) (o-クレゾール) ガリウム、ビス (2-メチル-8-キノリナート) (1-ナフトラート) アルミニウム、ビス (2-メチル-8-キノリナート) (2-ナフトラート) ガリウム等が挙げられるが、これらに限定されるものではない。

[0025]

また、含窒素五員環誘導体は、オキサゾール、チアゾール、オキサジアゾール、チアジアゾールもしくはトリアゾール誘導体が好ましい。具体的には、2, 5-ビス (1-フェニル) - 1, 3, 4-オキサゾール、ジメチルPPOP、2, 5-ビス (1-フェニル) - 1, 3, 4-チアゾール、2, 5-ビス (1-フェニル) - 1, 3, 4-オキサジアゾール、2- (4'-tert-ブチルフェニル) - 5- (4"-ビフェニル) 1, 3, 4-オキサジアゾール、2, 5-ビス (1-ナフチル) - 1, 3, 4-オキサジアゾール、1, 4-ビス [2- (5-フェニルオキサジアゾール)] ベンゼン、1, 4-ビス [2- (5-フェニルオキサジアゾール) - 4-tert-ブチルベンゼン]、2- (4'-tert-ブチルフェニル) - 5- (4"-ビフェニル) - 1, 3, 4-チアジアゾール、

(11)

JP 3958903 B2 2007.10.31

2, 5-ビス (1-ナフチル) -1, 3, 4-チアジアゾール、1, 4-ビス [2-(5-フェニルチアジアゾール)] ベンゼン、2-(4'-tert-ブチルフェニル)-5-(4"-ビフェニル)-1, 3, 4-トリアゾール、2, 5-ビス (1-ナフチル)-1, 3, 4-トリアゾール、1, 4-ビス [2-(5-フェニルトリアゾール)] ベンゼン等が挙げられるが、これらに限定されるものではない。

本発明においては、発光層と電極との間に無機化合物層を電荷注入性向上のために設けてもよい。このような無機化合物層としては、アルカリ金属化合物 (フッ化物、酸化物など)、アルカリ土類金属化合物などがあり、具体的には LiF 、 Li_2O 、 RaO 、 SrO 、 BaF_2 、 SrF_2 などが挙げられる。

【0026】

有機EL素子の陰極に使用される導電性材料としては、4 eV より大きな仕事関数を持つものが適しており、炭素、アルミニウム、バナジウム、鉄、コバルト、ニッケル、タングステン、銀、金、白金、パラジウム等およびそれらの合金、ITO 基板、NESA 基板に使用される酸化スズ、酸化インジウム等の酸化金属、さらにはポリチオフェンやポリビロール等の有機導電性樹脂が用いられる。陰極に使用される導電性物質としては、4 eV より小さな仕事関数を持つものが適しており、マグネシウム、カルシウム、鋳、鉛、チタニウム、イットリウム、リチウム、ルテニウム、マンガン、アルミニウム等およびそれらの合金が用いられるが、これらに限定されるものではない。合金としては、マグネシウム/銀、マグネシウム/インジウム、リチウム/アルミニウム等が代表例として挙げられるが、これらに限定されるものではない。合金の比率は、蒸着源の温度、雰囲気、真空度等により制御され、適切な比率に選択される。陽極および陰極は、必要があれば二層以上の層構成により形成されていてもよい。

【0027】

有機EL素子では、効率良く発光させるために、少なくとも一方の面は素子の発光波長領域において充分透明にすることが望ましい。また、基板も透明であることが望ましい。透明電極は、上記の導電性材料を使用して、蒸着やスパッタリング等の方法で所定の透光性が確保するように設定する。発光面の電極は、光透過率を10%以上にすることが望ましい。基板は、機械的、熱的強度を有し、透明性を有するものであれば限定されるものではないが、ガラス基板および透明性樹脂フィルムがある。透明性樹脂フィルムとしては、ポリエチレン、エチレン-酢酸ビニル共重合体、エチレン-ビニルアルコール共重合体、ポリプロピレン、ポリスチレン、ポリメタクリレート、ポリ塩化ビニル、ポリビニルアルコール、ポリビニルブチラール、ナイロン、ポリエーテルエーテルケトン、ポリサルホン、ポリエーテルサルホン、テトラフルオロエチレン-パーフルオロアルキルビニルエーテル共重合体、ポリビニルフルオライド、テトラフルオロエチレン-エチレン共重合体、テトラフルオロエチレン-ヘキサフルオロプロピレン共重合体、ポリクロロトリフルオロエチレン、ポリビニルフルオライド、ポリエステル、ポリカーボネート、ポリウレタン、ポリイミド、ポリエーテルイミド、ポリイミド、ポリプロピレン等が挙げられる。

【0028】

本発明の有機EL素子は、温度、湿度、雰囲気等に対する安定性の向上のために、素子の表面に保護層を設けたり、シリコンオイル、樹脂等により素子全体を保護することも可能である。

有機EL素子の各層の形成は、真空蒸着、スパッタリング、プラズマ、イオンプレーティング等の乾式成膜法やスピンコーティング、ディッピング、フローコーティング等の湿式成膜法のいずれの方法を適用することができる。膜厚は特に限定されるものではないが、適切な膜厚に設定する必要がある。膜厚が厚すぎると、一定の光出力を得るために大きな印加電圧が必要になり効率が悪くなる。膜厚が薄すぎるとピンホール等が発生して、電界を印加しても充分な発光強度が得られない。通常の膜厚は5 nmから10 μm の範囲が適しているが、10 nmから0.2 μm の範囲がさらに好ましい。

【0029】

(12)

JP 3998903 B2 2007.10.31

湿式成膜法の場合、各層を形成する材料を、エタノール、クロロホルム、テトラヒドロフラン、ジオキサン等の適切な溶媒に溶解または分散させて薄膜を形成するが、その溶媒はいずれであっても良い。また、いずれの有機薄膜層においても、成膜性向上、膜のピンホール防止等のため適切な樹脂や添加剤を使用しても良い。使用の可能な樹脂としては、ポリスチレン、ポリカーボネート、ポリアリレート、ポリエステル、ポリアミド、ポリウレタン、ポリスルホン、ポリメチルメタクリレート、ポリメチルアクリレート、セルロース等の絶縁性樹脂およびそれらの共重合体、ポリ-N-ビニルカルbazル、ポリシラン等の光導電性樹脂、ポリチオフェン、ポリピロール等の導電性樹脂を挙げられる。また、添加剤としては、酸化防止剤、紫外線吸収剤、可塑剤等を挙げられる。

【0030】

19

本発明の有機EL素子は、例えば壁掛けテレビのフラットパネルディスプレイ等の平面発光体、複写機、プリンター、液晶ディスプレイのバックライト又は計器類等の光源、表示板、標識灯等に利用できる。

【0031】

【実施例】

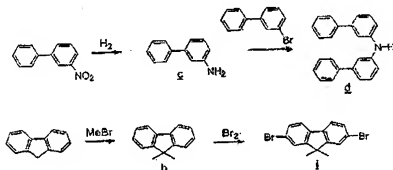
以下、本発明を合成例及び実施例に基づいてさらに詳細に説明する。

合成例1（化合物（A-2））

中間体d、中間体h、中間体iの反応経路を以下に示す。

【化7】

20



30

【0032】

中間体cの合成

3-ニトロジフェニル13g（65mmol）のエタノール75ミリリットル懸濁液中に、Pd/C（7.5%）1gを仕込み、30℃以下にて水素を吹き込みながら7時間反応させた。反応液を濾過し、Pd/C除去した後、溶媒を減圧留まし、目的とする中間体c 10.8gを得た（収率9.8%）。

中間体dの合成

40

アルゴン気流下、冷却管付き300ミリリットル三口フラスコ中に、中間体c 6.8g（40mmol）、3-ブロモフェニル9.2g（40mmol）、トリス（ジベンジリデンアセトン）ジバリウム1.1g（1.5mmol）、トリ-*o*-トルイルホスフィン0.72g（3mmol）、*tert*-ブトキシナトリウム3.8g（40mmol）、乾燥トルエン100ミリリットルを加えた後、100℃にて一晩加熱撹拌した。反応終了後、析出した結晶を濾取し、メタノール100ミリリットルにて洗浄し、中間体d 11.8g（収率90%）を得た。

【0033】

中間体hの合成

アルゴン気流下、500ミリリットル三口フラスコ中に、フルオレン2.2g（0.13

50

(13)

JP 3958903 B2 2007.10.31

mmol)、乾燥THF100ミリリットルを仕込み-78℃に冷却した。ここに、n-ブチルチウム(2.6Mヘキサン)120ミリリットル(0.32mol)を滴下した。同温にて1時間攪拌した後、メチルプロピド28g(0.3mol)/THF60ミリリットル溶液を-78℃にて滴下した。その後、徐々に室温に戻し、室温にて一晩攪拌した。反応終了後、反応液を水1リットルに注入し、IPE抽出、飽和食塩水洗浄後、無水硫酸マグネシウムで乾燥留置した。残さをカラムクロマトグラフィー(シリカゲル、展開溶媒：ヘキサン)にて精製し、中間体h25g(収率98%)を得た。

中間体iの合成

透光した1リットル三口フラスコ中に、中間体h9.7g(50mmol)、クロロホルム100ミリリットル、FeCl₃0.2gを仕込んだ。次いで、臭素24g(0.15mol)を0℃にて滴下した。その後、室温で一晩反応させた。反応終了後、析出物を濾取、水洗、エタノールを洗浄後、加熱乾燥し目的とする中間体i15gを得た(収率85%)。

化合物(A-2)の合成

アルゴン気流下、冷却管付き300ミリリットル三口フラスコ中に、中間体i3.5g(10mmol)、中間体d6.4g(20mmol)、トリス(ジベンジリデンアセトン)ジブタジウム0.27g(1.5mol%)、トリ-オ-トルイルホスフィン0.18g(3mol%)、t-ブトキシナトリウム1.9g(20mmol)、乾燥トルエン100ミリリットルを加えた後、100℃にて一晩加熱攪拌した。反応終了後、析出した結晶を濾取し、メタノール100ミリリットルにて洗浄し、黄色粉末6.6gを得た。このものは、NMR、IR及びFD-MS(フィールドイオンマスマススペクトル)の測定により、化合物(A-2)と同一とされた(収率80%)。

[0034]

実施例1

2.5mm×7.5mm×1.1mm厚のITO透明電極付きガラス基板(ジオマテック社製)をイソプロピルアルコール中で超音波洗浄を5分間行なった後、UVオゾン洗浄を30分間行なった。洗浄後の透明電極ライン付きガラス基板を真空蒸着装置の基板ホルダーに装着し、まず透明電極ラインが形成されている側の面上に、前記透明電極を覆うようにして膜厚60nmのN,N'-ビス(N,N'-ジフェニル-4-アミノフェニル)-N,N'-ジフェニル-4,4'-ジアミノ-1,1'-ビフェニル膜(以下、TPD232膜)を成膜した。このTPD232膜は、正孔注入層として機能する。次に、TPD232膜上に膜厚20nmの上記した正孔輸送性化合物(A-2)を成膜した。この化合物(A-2)膜は正孔輸送層として機能する。さらに、化合物(A-2)膜上に膜厚40nmのトリス(8-キノリノール)アルミニウム膜(以下、Alq膜)を成膜した。このAlq膜は、発光層として機能する。この後Li(Li源：サエスゲッター社製)とAlqを二元蒸着させ、電子注入層(陰極)としてAlq:Li膜を膜厚20nmで形成した。このAlq:Li膜上に金属Alを蒸着させ金属陰極を形成し有機EL素子を作製した。

この素子は直流電圧6Vで発光輝度153cd/m²、最大発光輝度50000cd/m²、発光効率3.2cd/Aの青色発光が得られた。また、耐熱保存試験として、100℃の環境で500時間保存した。試験前と同様に直流電圧6Vを印加したところ、初期の輝度に対し98%の輝度を示し、輝度保持率98%であった。

[0035]

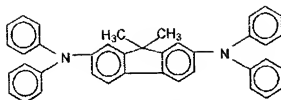
比較例1

実施例1において、化合物(A-2)の代わりに、下記化合物TPAF(ガラス転移温度100℃未満)

[化8]

(14)

JP 398903 B2 2007.10.31



を使用したことを除き同様にして、有機EL素子を作製し、直流電圧5Vで、発光輝度、
発光効率を測定し、発光色を観察し、さらに耐熱性テストとして85℃の温度下に500
時間保存した後に、初期輝度からの輝度保持率を測定した。その結果を表1に示す。

【0036】

【表1】

表 1

	化合物 の種類	電圧 (V)	発光輝度 (cd/m ²)	発光効率 (cd/A)	発光色	輝度保持 率(%)
比較例1	TPAF	5	150	2.5	緑	56

【0037】

表1に示したように、本発明の新規アリールアミン化合物を利用した有機EL素子は、
発光輝度、発光効率が高く、耐熱性に優れている。これは、本発明の新規アリールアミン
化合物のガラス転移温度が100℃以上と高く、発光層と相互作用しないためである。

【0038】

【発明の効果】

以上、詳細に説明したように、本発明の新規アリールアミン化合物を利用した有機エ
レクトロルミネッセンス素子は、高輝度で、耐熱性が高く、長寿命で、正孔輸送性が優れ高
発光効率である。

このため、本発明の有機エレクトロルミネッセンス素子は、壁掛テレビの平面発光体や
ディスプレイのバックライト等の光源として有用である。

(15)

JP 3998903 B2 2007.10.31

フロントページの続き

(56)参考文献 特開2000-016973 (JP, A)

特開平07-287408 (JP, A)

特開平11-184119 (JP, A)

特開平10-104861 (JP, A)

特開昭63-094249 (JP, A)

特開平11-154594 (JP, A)

特開平11-185965 (JP, A)

特開平11-185966 (JP, A)

特開平11-167992 (JP, A)

(58)調査した分野(Int.Cl., DB名)

G06C 211/61

G06C 59/00

REGISTRY (STN)

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

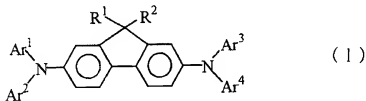
CLAIMS

(57) [Claim(s)]

[Claim 1]

The new arylamine compound expressed with the following general formula (1).

[Formula 1]



(R1 and R2 express independently the aryloxy group of the carbon atomic numbers 6-40 which are not permuted / the arylated alkyl radical of the carbon atomic numbers 7-40 which are not permuted / the aryl group of the carbon atomic numbers 6-40 which are not permuted / the alkoxy group of the carbon atomic numbers 1-30 which are not permuted / the alkyl group of the carbon atomic numbers 1-30 which are not permuted / a permutation or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or /, among a formula, respectively.)

Ar1 -Ar4 Independently, the aryl group of the carbon atomic numbers 6-40 which are not permuted [a permutation or] is expressed, and even if respectively the same, you may differ, respectively. However, Ar1 -Ar4 At least two are m-biphenyl which is not permuted [a permutation or] inside, and the remainder is a biphenyl which is not permuted [a permutation or].

[Claim 2]

It sets to the new arylamine compound of said general formula (1), and is Ar1. And Ar3 m-biphenyl which is not permuted [a permutation or] and Ar2 And Ar4 New arylamine compound according to claim 1 characterized by being the biphenyl which is not permuted [a permutation or].

[Claim 3]

The charge of organic electroluminescent element material which consists of a new arylamine compound expressed with a general formula (1) according to claim 1.

[Claim 4]

The organic electroluminescent element which is an organic electroluminescent element which has an organic compound layer in inter-electrode [of a pair], and is characterized by this organic compound layer containing the charge of organic electroluminescent element material according to claim 3.

[Claim 5]

The organic electroluminescent element according to claim 4 to which said organic compound layer is

characterized by being a luminous layer or an electron hole transportation layer.

[Claim 6]

The organic electroluminescent element which is an organic electroluminescent element which has an organic compound layer in inter-electrode [of a pair], and is characterized by having the layer in which this organic compound layer contains the charge of organic electroluminescent element material according to claim 3 and luminescent material.

[Translation done.]

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.*** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

this invention -- a new arylamine compound and an organic electroluminescent element -- being related -- especially -- high brightness -- it is -- thermal resistance -- high -- long lasting -- electron hole transportability -- excelling -- high -- it is related with the new arylamine compound which realizes luminous efficiency organic electroluminescent element and it.

[0002]

[Description of the Prior Art]

The organic electroluminescence (EL) component which used the organic substance is used as the light source of the flat-surface illuminant of wall tapestry television, the back light of a display, etc., and development is performed briskly.

the electroluminescence phenomenon of an organic material will be observed by Pope and others (Pope) with an anthracene single crystal in 1963 (J. Chem.Phys.38 (1963) 2042) -- HERUFURIIHI (Helfinch) and Schneider (Schneider) have succeeded in observation of comparatively strong injection EL by using a solution electrode system with sufficient injection efficiency in 1965 (Phys.Rev.Lett.14 (1965) 229). As reported since then, research which formed the organic luminescence matter with the conjugate organic host substance and the organic activator conjugate [with the condensation benzene ring] is done. Naphthalene, an anthracene, a phenanthrene, tetracene, a pyrene, A benzopyrene, a chrysene, picene, a carbazole, a fluorene, a biphenyl, Terphenyl, triphenylene oxide, dihalo biphenyl, trans-stilbene and 1, and 4-diphenyl butadiene etc. was shown as an example of an organic host substance, and an anthracene, tetracene, pentacene, etc. were mentioned as an example of an activator. However, each of these organic luminescence matter existed as a monolayer with the thickness exceeding 1 micrometer, and needed high electric field for luminescence. For this reason, research of the thin film by the vacuum deposition method was advanced (for example, Thin Solid Films 94 (1982) 171). . However, although thin-film-izing was effective in reduction of driver voltage, it did not come to obtain the component of high brightness of practical use level.

Then, tongues (Tang) devised the EL element which carried out the laminating of the two very thin film (an electron hole transportation layer and luminous layer) with vacuum deposition between an anode plate and cathode, and realized high brightness by low driver voltage (Appl.Phys.Lett.51 (1987) 913 or U.S. Pat. No. 4356429 number). Then, as a result of furthering development of the organic compound used for an electron hole transportation layer and a luminous layer for about ten years, the life and luminous efficiency of utilization level were attained. Consequently, as for the organic EL device, utilization is started from the display of a car stereo and a cellular phone etc.

[0003]

However, in the practical use side, luminescence brightness, the endurance of degradation with the passage of time to long duration use, etc. are not enough, and the further improvement is called for. It is [as opposed to / when the application to a full color display etc. is considered especially / each color of

R, G, and B] 300 cd/m². Reaching the reduction-by-half life of thousands of hours or more by the above high brightness is called for. Since the energy gap of a luminous layer was as large as 2.8eV or more for blue luminescence realizing this and carrying out blue luminescence especially of the difficult thing and the energy barrier in the case of the hole injection between an electron hole transportation layer and a luminous layer was large, the field strength impressed to an interface was large, in the conventional electron hole transportation layer, a hole injection was not completed in stability but amelioration was called for.

Moreover, when premised on carrying out vehicle loading of the organic EL device, it is pointed out that a problem is in elevated-temperature shelf-life ability 100 degrees C or more. Although it is pointed out that glass transition temperature is low in the conventional electron hole transportation layer also in this case and it tended to correspond only by improving this at 100 degrees C or more, good shelf-life ability [in / it is inadequate and / an elevated temperature] was not yet realized. Furthermore, exciplex was generated as an interaction of an electron hole transportation layer and a luminous layer, and there was also a problem that the brightness of a component deteriorated.

[0004]

[Problem(s) to be Solved by the Invention]

what was made in order that this invention might solve the aforementioned technical problem -- it is -- high brightness -- it is -- thermal resistance -- high -- long lasting -- electron hole transportability -- excelling -- high -- it aims at offering the new arylamine compound which realizes luminous efficiency organic electroluminescent element and it.

[0005]

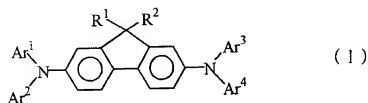
[Means for Solving the Problem]

this invention persons came to complete header this invention for the brightness of an organic electroluminescent element, thermal resistance, and a life improving, if the new arylamine compound which has specific structure is added in an organic compound layer, and electron hole transportability improving further, and becoming high luminous efficiency, as a result of repeating research wholeheartedly that the organic electroluminescent element (the following, organic EL device) which has the aforementioned desirable property should be developed.

[0006]

That is, this invention offers the new arylamine compound expressed with the following general formula (1).

[Formula 2]



[0007]

(R1 and R2 express independently the aryloxy group of the carbon atomic numbers 6-40 which are not permuted [the arylated alkyl radical of the carbon atomic numbers 7-40 which are not permuted / the aryl group of the carbon atomic numbers 6-40 which are not permuted / the alkoxy group of the carbon atomic numbers 1-30 which are not permuted / the alkyl group of the carbon atomic numbers 1-30 which are not permuted / a permutation or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or / among a formula, respectively.)

Ar1 -Ar4 Independently, the aryl group of the carbon atomic numbers 6-40 which are not permuted [a

permutation or] is expressed, and even if respectively the same, you may differ, respectively. However, Ar1 -Ar4 At least two are m-biphenyl which is not permuted [a permutation or] inside, and the remainder is a biphenyl which is not permuted [a permutation or].

Moreover, this invention is an organic EL device which has an organic compound layer in inter-electrode [of a pair], and provides the charge of organic EL device material which consists of a new arylamine compound expressed with a general formula (1), and a list also with the organic EL device characterized by this organic compound layer containing said charge of organic EL device material.

[0008]

[Embodiment of the Invention]

The new arylamine compound of this invention is expressed with the above-mentioned general formula (1).

It sets to a general formula (1) and is R1. And R2 The aryloxy group of the carbon atomic numbers 6-40 which are not permuted [the arylated alkyl radical of the carbon atomic numbers 7-40 which are not permuted / the aryl group of the carbon atomic numbers 6-40 which are not permuted / the alkoxy group of the carbon atomic numbers 1-30 which are not permuted / the alkyl group of the carbon atomic numbers 1-30 which are not permuted / a permutation or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or] is expressed independently, respectively.

As an alkyl group, for example as an alkoxy group, methyl, ethyl, n-propyl, iso-propyl, etc. As methoxy, ethoxy **, and an aryl group, as an arylated alkyl radical, phenyl, a biphenyl, naphthyl, etc. Benzyl, alpha-methylbenzyl, alpha-ethyl benzyl, alpha, and alpha-dimethylbenzyl, As an aryloxy group, 4-methylbenzyl, 4-ethyl benzyl, 2-tert-butylbenzyl, 4-n-octylbenzyl, naphthyl methyl, diphenyl methyl, etc. Phenoxy, naphthyl oxy one, and ANSUKURU oxy-*** pyrenyl oxy-*** biphenyl oxy-*** fluoran thenyl oxy-*** KURISE nil oxy-*** peri RENIRU oxy-*** are mentioned.

[0009]

As a substituent of each [these] radical, for example Moreover, a fluorine atom, a chlorine atom, Halogen atoms, such as a bromine atom and iodine atom, methyl, ethyl, n-propyl, Alkyl groups, such as iso-propyl, methoxy, the alkoxy group of ethoxy **, Arylated alkyl radicals, such as aryloxy groups, such as phenoxy, benzyl, phenethyl, and phenylpropyl, A nitro group, a cyano group, a dimethylamino radical, a dibenzylamino radical, a diphenylamino radical, Heterocycle radicals, such as aryl groups, such as permutation amino groups, such as a morpholino group, a phenyl group, a toluyl radical, a biphenyl radical, a naphthyl group, an anthryl radical, and a pyrenyl radical, a pyridyl radical, a thienyl group, a furil radical, a quinolyl radical, and a cull PAZORIRU radical, etc. are mentioned.

[0010]

Moreover, it sets to a general formula (1) and is Ar1 -Ar4. Independently, the aryl group of the carbon atomic numbers 6-40 which are not permuted [a permutation or] is expressed, and even if respectively the same, you may differ, respectively.

As an aryl group, a phenyl group, a toluyl radical, a biphenyl radical, a naphthyl group, an anthryl radical, a pyrenyl radical, etc. are mentioned, for example.

It sets to a general formula (1) and is Ar1 -Ar4. At least two are m-biphenyl which is not permuted [a permutation or] inside, and the remainder is a biphenyl which is not permuted [a permutation or].

[0011]

Ar1 -Ar4 As a substituent in the radical to express For example, halogen atoms, such as a fluorine atom, a chlorine atom, a bromine atom, and iodine atom, Alkyl groups, such as methyl, ethyl, n-propyl, and iso-propyl, Aryloxy groups, such as methoxy, an alkoxy group of ethoxy **, and phenoxy, Arylated alkyl radicals, such as benzyl, phenethyl, and phenylpropyl, A nitro group, a cyano group, a dimethylamino radical, a dibenzylamino radical, a diphenylamino radical, Heterocycle radicals, such as aryl groups, such as permutation amino groups, such as a morpholino group, a phenyl group, a toluyl radical, a biphenyl radical, a naphthyl group, an anthryl radical, a pyrenyl radical, and a fluoran thenyl radical, a pyridyl radical, a thienyl group, a furil radical, a quinolyl radical, and a cull PAZORIRU radical, etc. are mentioned.

Furthermore, as an aryl substituent in an aryl permutation biphenyl, a phenyl group, a biphenyl radical, a

terphenyl radical, a naphthyl group, an anthryl radical, a fluorenyl group, etc. are mentioned, for example.

Ar1 And Ar3 m-biphenyl which is not permuted [a permutation or] and Ar2 And Ar4 It is desirable that it is the biphenyl which is not permuted [a permutation or].

[0012]

The organic EL device of this invention is an organic EL device which has an organic compound layer in inter-electrode [of a pair], and this organic compound layer contains the charge of organic EL device material of said new arylamine compound.

It is desirable that said organic compound layer is a luminous layer or an electron hole transportation layer. Moreover, it is also desirable to have the layer in which said organic compound layer contains the charge of organic EL device material and luminescent material of said new arylamine compound.

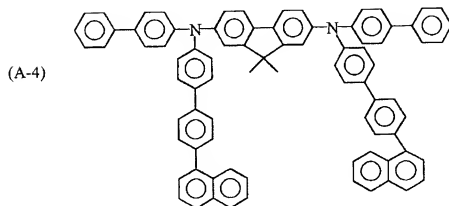
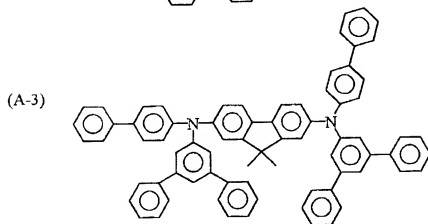
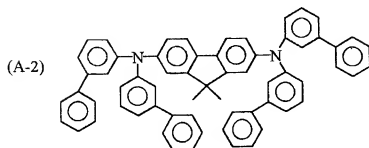
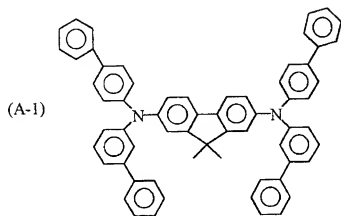
it is because at least non-***** which it has a high glass transition point that the brightness of an organic EL device, thermal resistance, a life, and luminous efficiency improve, and it cannot interact with luminescent material easily when this arylamine compound is excellent in electron hole transportability and a hole injection is possible for stability, and is produced by the interaction will be avoided if an organic compound layer boils said new arylamine compound further at least and it is made to contain.

[0013]

Although the example of representation of a general formula (1) is illustrated in the new arylamine compound of this invention below, this invention is not limited to these examples of representation.

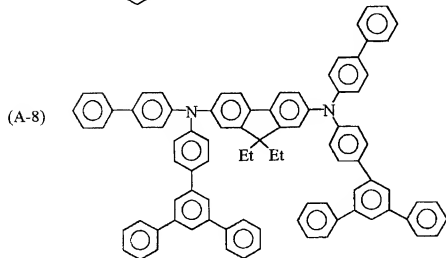
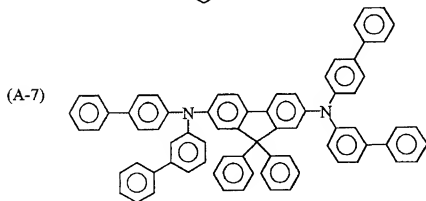
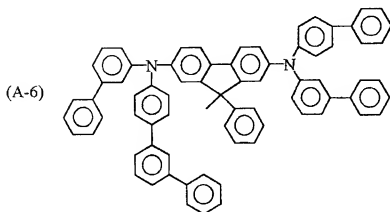
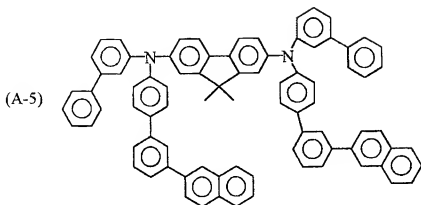
[0014]

[Formula 3]



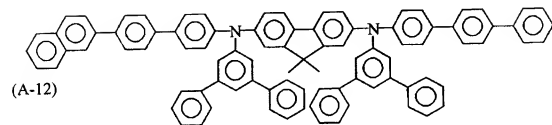
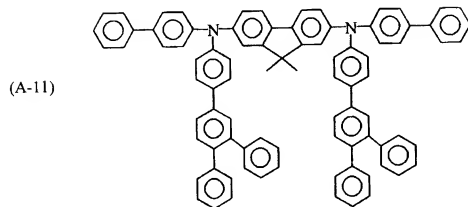
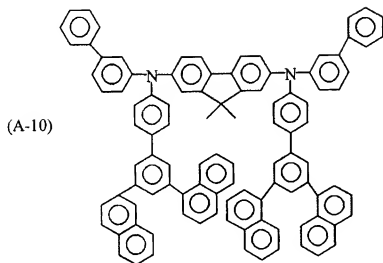
[0015]

[Formula 4]



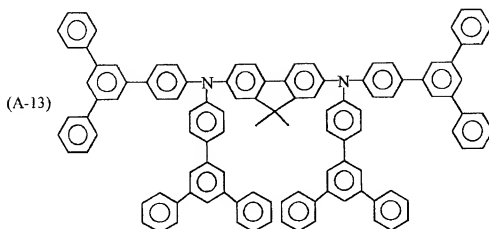
[0016]

[Formula 5]



[0017]

[Formula 6]



[0018]

The organic EL device of this invention is a component in which the monolayer or the multilayer organic compound layer was formed between an anode plate and cathode. In the case of the monolayer mold, the luminous layer is prepared between an anode plate and cathode. A luminous layer contains luminescent material, and in order to make the electron hole which was poured in from the anode plate in addition to it, or the electron poured in from cathode convey to luminescent material, it may contain a hole-injection ingredient or an electron injection ingredient. However, as for luminescent material, it is desirable to have very high fluorescence quantum efficiency, high electron hole transport capacity, and electronic transport capacity, and to form a uniform thin film. The organic EL device of a multilayer mold has some which carried out the laminating with the multilayer configuration of (an anode plate / hole injection layer / luminous layer / cathode), (an anode plate / luminous layer / electronic injection layer / cathode), and (an anode plate / hole injection layer / luminous layer / electronic injection layer / cathode).

[0019]

In addition to the new arylamine compound of this invention, the further well-known luminescent material, a doping ingredient, a hole-injection ingredient, and an electron injection ingredient can also be used for a luminous layer if needed. As a desirable usage of this new arylamine compound, it adds at 0.5 - 100 % of the weight of concentration in the layer of either a luminous layer, an electronic injection layer, an electron hole transportation layer or a hole injection layer. It is 50 - 100 % of the weight of concentration still more preferably.

An organic EL device can prevent the fall of the brightness by quenching, or a life by making it multilayer structure. If there is need, it can be used combining luminescent material, other doping ingredients, a hole-injection ingredient, or an electron injection ingredient. Moreover, improvement in luminescence brightness or luminous efficiency and luminescence of red or white can also be obtained with other doping ingredients. Moreover, a hole injection layer, a luminous layer, and an electronic injection layer may be formed of the lamination more than a bilayer, respectively. In the case of a hole injection layer, in that case, the layer which conveys [the layer which pours in an electron hole from an electrode] an electron hole for an electron hole from a hole injection layer and a hole injection layer to a reception luminous layer is called an electron hole transportation layer. Similarly, in the case of an electronic injection layer, the layer which conveys [the layer which pours in an electron from an electrode] an electron for an electron from an electronic injection layer and an electronic injection layer to a reception luminous layer is called an electron transport layer. These each class is used by each factor, such as adhesion with the energy level of an ingredient, thermal resistance, an organic compound

layer, or a metal electrode, choosing it.

[0020]

As the luminescent material which can be used for an organic compound layer with said new arylamine compound, or a host ingredient, there is condensed multi-ring aromatic series, for example, an anthracene, naphthalene, a phenanthrene, a pyrene, tetracene, pentacene, coronene, a chrysene, a fluorescein, perylene, rubrene, and those derivatives exist. Non [furthermore, / phthal perylene, naphthalene perylene, and peri non, / phthal peri] Naphthalene peri non, a diphenyl butadiene, a tetra-phenyl butadiene, A coumarin, oxadiazole, aldazine, bis-benzoKISAZORIN, Bis-styryl, pyrazine, a cyclopentadiene, a quinoline metal complex, An amino quinoline metal complex, a benzoquinoline metal complex, an imine, diphenylethylene, Although a vinyl anthracene, a diamino carbazole, a pyran, thiopyran, poly methine, merocyanine, an imidazole chelation oxy-NOIDO compound, Quinacridone, rubrene, a stilbene system derivative, a fluorochrome, etc. are mentioned, it is not limited to these.

[0021]

The compound which has the capacity to convey an electron hole, as a well-known hole-injection ingredient, has the hole-injection effectiveness which was excellent to the hole-injection effectiveness, the luminous layer, or luminescent material from an anode plate, and prevented migration into the electronic injection layer or electron injection ingredient of an exciton generated by the luminous layer, and was excellent in the thin film organization potency force is desirable. Specifically A phthalocyanine derivative, a naphthalocyanine derivative, a porphyrin derivative, Oxazole, oxadiazole, triazole, an imidazole, imidazolone, Imidazole thione, pyrazoline, a pyrazolone, a tetrahydro imidazole, Oxazole, oxadiazole, a hydrazone, an acyl hydrazone, The poly aryl alkane, a stilbene, a butadiene, a benzidine mold triphenylamine, a styryl amine mold triphenylamine, a diamine mold triphenylamine, etc., Although polymeric materials, such as those derivatives and a polyvinyl carbazole, polysilane, and a conductive polymer, are mentioned, it is not limited to these.

[0022]

The still more effective hole-injection ingredient in the well-known hole-injection ingredient which can be used in the organic EL device of this invention is the third class amine derivative of aromatic series, or a phthalocyanine derivative.

The example of the third class amine derivative of aromatic series A triphenylamine, a tritolyl amine, A tolyl diphenylamine, N, N'-diphenyl-N, N' - (3-methylphenyl) -1, the 1'-biphenyl -4, 4'-diamine, N, N, N', N' - (4-methylphenyl) -1, the 1'-phenyl -4, 4'-diamine, N, N, N', N' - (4-methylphenyl) -1, the 1'-biphenyl -4, 4'-diamine, N and N' - diphenyl-N and N' - dinaphthyl -1 and 1' -- the 1'-biphenyl -4 and 4' - diamine -- N, N'-(methylphenyl)-N, N' -(4-n-butylphenyl)- A phenanthrene -9, 10-diamine, Although it is oligomer or a polymer with the third class amines frame of such aromatic series, such as an N and N-bis(4-G 4-tolylamino phenyl)-4-phenyl-cyclohexane, it is not limited to these.

As for the example of a phthalocyanine (Pc) derivative, ***** is not limited to these with a phthalocyanine derivative and naphthalocyanine derivatives, such as H2 Pc, CuPc, CoPc, NiPc, ZnPc, PdPc, FePc, MnPc, ClAlPc, ClGaPc, ClInPc, ClSnPc, Cl2 SiPc, (HO) AlPc, (HO) GaPc, VOPc, TiOPc, MoOPc, and GaPc-O-GaPc.

[0023]

The compound which has the capacity to convey an electron, as a well-known electron injection ingredient, has the electron injection effectiveness which was excellent to the electron injection effectiveness, the luminous layer, or luminescent material from cathode, and prevented migration to the hole injection layer of the exciton generated by the luminous layer, and was excellent in the thin film organization potency force is desirable. concrete -- full -- me -- non, although anthra quinodimethan, diphenylquinone, thiopyran dioxide, oxazole, oxadiazole, triazole, an imidazole, perylene tetracarboxylic acid, deflection ORENIRIDEN methane, anthra quinodimethan, anthrones, etc. and those derivatives are mentioned, it is not limited to these. Moreover, charge impregnation nature can also be raised by adding the electronic acceptance matter into a hole-injection ingredient, and adding the electron-donative matter into an electron injection ingredient.

[0024]

In the organic EL device of this invention, a still more effective well-known electron injection ingredient is a metal complex compound or a nitrogen-containing five membered ring derivative.

The example of a metal complex compound 8-hydroxyquinolate lithium, Bis(8-hydroxyquinolate) zinc, bis(8-hydroxyquinolate)copper, Bis(8-hydroxyquinolate) manganese, tris(8-hydroxyquinolate)aluminium, Tris (2-methyl-8-hydroxyquinolate) aluminum, A tris (8-hydroxyquinolate) gallium, bis(10-hydroxy benzo[h] quinolate) beryllium, Bis(10-hydroxy benzo[h] quinolate) zinc, a bis(2-methyl-8-quinolate) chloro gallium, Although a bis(2-methyl-8-quinolate) (o-cresolate) gallium, bis(2-methyl-8-quinolate) (1-naphth RATO) aluminum, a bis(2-methyl-8-quinolate) (2-naphth RATO) gallium, etc. are mentioned It is not limited to these.

[0025]

Moreover, a nitrogen-containing 5 member derivative has oxazole, a thiazole, oxadiazole, thiadiazole, or a desirable triazole derivative. Specifically, it is 2 and 5-screw (1-phenyl). - 1, 3, 4-oxazole, Dimethyl POPOP, 2, 5-screw (1-phenyl) - 1, 3, 4-thiazole, 2, 5-screw (1-phenyl) - 1, 3, 4-oxadiazole, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl) 1, 3, 4-oxadiazole, 2, 5-bis(1-naphthyl)-bis[1, 3, 4-oxadiazole, 1, and 4-] [2- (5-phenyl oxadiazolyl)] benzene, 1, 4-screw [2-(5-phenyl oxadiazolyl)-4-tert-butylbenzene], 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)- 1, 3, and 4-thiadiazole -- 2, 5-bis(1-naphthyl)-bis[1, 3, 4-thiadiazole, 1, and 4-] [2- (5-phenyl thiadiazolyl)] benzene, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-, although 1, 3, 4-triazole, 2, and 5-bis(1-naphthyl)-bis[1, 3, 4-triazole, 1, and 4-] [2- (5-phenyl triazolyl)] benzene etc. is mentioned It is not limited to these.

In this invention, an inorganic compound layer may be prepared between a luminous layer and an electrode for the improvement in charge injectional. as such an inorganic compound layer -- alkali metal compounds (a fluoride, oxide, etc.), an alkaline-earth-metal compound, etc. -- it is -- concrete -- LiF, Li2 O, RaO, SrO and BaF2, and SrF2 etc. -- it is mentioned.

[0026]

What has a bigger work function than 4eV as a conductive ingredient used for the anode plate of an organic EL device is suitable, and organic conductive resin, such as the poly thiophene and polypyrrole, is used for gold oxide groups, such as tin oxide used for those alloys, such as carbon, aluminum, vanadium, iron, cobalt, nickel, a tungsten, silver, gold, platinum, and palladium, and an ITO substrate, and a NESA substrate, and indium oxide, and a pan. Although what has a work function smaller than 4eV as conductive matter used for cathode is suitable and those alloys, such as magnesium, calcium, tin, lead, titanium, an yttrium, a lithium, a ruthenium, manganese, and aluminum, are used, it is not limited to these. As an alloy, although magnesium/silver, magnesium/indium, a lithium/aluminum, etc. are mentioned as an example of representation, it is not limited to these. The ratio of an alloy is controlled by the temperature of the source of vacuum evaporation, the ambient atmosphere, a degree of vacuum, etc., and is chosen as a suitable ratio. As long as an anode plate and cathode have the need, they may be formed of the lamination more than a bilayer.

[0027]

In order to make light emit efficiently in an organic EL device, as for one [at least] field, it is desirable to make it transparence enough in the luminescence wavelength field of a component. Moreover, the transparent thing of a substrate is desirable. The above-mentioned conductive ingredient is used for a transparent electrode, and it sets it up so that predetermined translucency may secure by approaches, such as vacuum evaporation and sputtering. As for the electrode of a luminescence side, it is desirable to make light transmittance 10% or more. Although a substrate is not limited if it has mechanical and thermal reinforcement and has transparency, it has a glass substrate and a transparency resin film. As a transparency resin film, polyethylene, an ethylene-vinylacetate copolymer, An ethylene-vinylalcohol copolymer, polypropylene, polystyrene, Polymethylmethacrylate, a polyvinyl chloride, polyvinyl alcohol, A polyvinyl butyral, nylon, a polyether ether ketone, the poly ape phone, A polyether ape phon, a tetrafluoroethylene-perfluoroalkyl vinyl ether copolymer, Polyvinyl fluoride, a tetrafluoroethylene-ethylene copolymer, A tetrafluoroethylene-hexafluoropropylene copolymer, polychlorotrifluoroethylene resin, poly vinylidene fluoride, polyester, a polycarbonate, polyurethane, polyimide, polyether imide, polyimide, polypropylene, etc. are mentioned.

[0028]

A protective layer is prepared on the surface of a component for the improvement of stability to temperature, humidity, an ambient atmosphere, etc., or the organic EL device of this invention can also protect the whole component with a silicone oil, resin, etc.

Formation of each class of an organic EL device can apply which approach of the wet forming-membranes methods, such as the dry type forming-membranes methods, such as vacuum deposition, sputtering, plasma, and ion plating, spin coating, dipping, and flow coating. Although especially thickness is not limited, it is necessary to set it as suitable thickness. If thickness is too thick, in order to obtain a fixed optical output, big applied voltage will be needed and effectiveness will worsen. If thickness is too thin, even if a pinhole etc. will occur and it will impress electric field, sufficient luminescence brightness is not obtained. The usual thickness has the still more desirable range of 0.2 micrometers from 10nm, although the range of 10 micrometers is suitable from 5nm.

[0029]

The solvent may be any, although suitable solvents, such as ethanol, chloroform, a tetrahydrofuran, and dioxane, are made to dissolve or distribute the ingredient which forms each class in the case of the wet forming-membranes method and a thin film is formed. Moreover, also in which organic thin film layer, suitable resin and a suitable additive may be used on a membrane formation disposition for pinhole prevention of the film etc. As possible resin of use, conductive resin, such as photoconductivity resin, such as insulating resin, such as polystyrene, a polycarbonate, polyarylate, polyester, a polyamide, polyurethane, polysulfone, polymethylmethacrylate, polymethyl acrylate, and a cellulose, and those copolymers, poly-N-vinylcarbazole, and polysilane, the poly thiophene, and polypyrrole, can be mentioned. Moreover, an antioxidant, an ultraviolet ray absorbent, a plasticizer, etc. can be mentioned as an additive.

[0030]

The organic EL device of this invention can be used for the light source of the back light of flat-surface illuminants, such as a flat-panel display of a flat TV, a copying machine, a printer, and a liquid crystal display, or instruments, the plotting board, a beacon light, etc.

[0031]

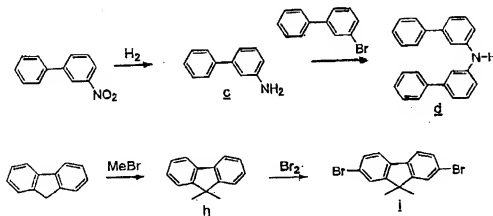
[Example]

Hereafter, this invention is further explained to a detail based on a synthetic example and an example.

The synthetic example 1 (compound (A-2))

The reaction path of intermediate field d, intermediate field h, and intermediate field i is shown below.

[Formula 7]



[0032]

Composition of intermediate field c

It was made to react for 7 hours, teaching Pd/C(7.5%) 1g and blowing hydrogen below 30 degrees C into the 3-nitro diphenyl 13g (65mmol) 75ml suspension of ethanol. Reaction mixture was filtered, and after carrying out Pd/C removal, intermediate-field c10.8g which carries out reduced pressure distilling off of the solvent, and is made into the purpose was obtained (98% of yield).

Composition of intermediate field d

They are intermediate-product c6.8g (40mmol), 3-BUOMO phenyl 9.2g (40mmol), and tris (JIBEN zylidene acetone) the bottom of an argon air current, and in a 300ml three necked flask with a cooling pipe. After adding JIPARAJIUMU1.1g (1.5-mol%), tree o-tolulyl phosphine 0.72g (three-mol%), t-butoxy sodium 3.8g (40mmol), and 100ml of desiccation toluene, heating **** was carried out at 100 degrees C overnight. The depositing crystal was separated after reaction termination, 100ml of methanols washed, and intermediate-field d11.8g (90% of yield) was obtained.

[0033]

Composition of intermediate field h

Fluorene 22g (0.13mmol) and 100ml of desiccation THF were taught the bottom of an argon air current, and into the 500ml three necked flask, and it cooled at -78 degrees C. 120ml (2.6M hexane) (0.32 mols) of n-butyl lithium was dropped here. After agitating in this ** for 1 hour, the 60ml solution of methyl bromide 28g (0.3 mols)/THF(s) was dropped at -78 degrees C. Then, it returned to the room temperature gradually and ****(ed) at the room temperature overnight. Reaction mixture was poured into 1l. of water after reaction termination, and desiccation distilling off was carried out with sulfuric anhydride gel, an expansion solvent: hexane) refined the residue, and intermediate-field h25g (98% of yield) was obtained.

Composition of intermediate field i

Into the 1l. three necked flask which shaded, intermediate-field h9.7g (50mmol), 100ml of chloroform, and FeCl₂ 0.2g were taught. Subsequently, 24g (0.15 mols) of bromines was dropped at 0 degree C. Then, it was made to react at a room temperature overnight. Intermediate-field i15g which carries out stoving of the precipitated crystal after washing separation, rinsing, and ethanol, and targets it was obtained after reaction termination (85% of yield).

Composition of a compound (A-2)

They are intermediate-product i3.5g (10mmol), intermediate-product d6.4g (20mmol), and tris (JIBEN zylidene acetone) the bottom of an argon air current, and in a 300ml three necked flask with a cooling pipe. After adding JIPARAJIUMU0.27g (1.5-mol%), tree o-tolulyl phosphine 0.18g (three-mol%), t-butoxy sodium 1.9g (20mmol), and 100ml of desiccation toluene, heating **** was carried out at 100 degrees C overnight. The depositing crystal was separated after reaction termination, 100ml of methanols washed, and 6.6g of yellow powder was obtained. This thing was identified the compound (A-2) by measurement of NMR, IR, and FD-MS (field desorption mass spectrum) (80% of yield).

[0034]

Example 1

After performing ultrasonic cleaning for the glass substrate with an ITO transparent electrode of 25mmx75mmx1.1mm thickness (JIOMA tick company make) for 5 minutes in isopropyl alcohol, UV ozone washing was performed for 30 minutes. the field top of the side in which the substrate electrode holder of a vacuum evaporation system is equipped with the glass substrate with transparent electrode Rhine after washing, and transparent electrode Rhine is formed first -- said transparent electrode -- a wrap -- like -- carrying out -- N of 60nm of thickness, and N' - bis(N and N'-diphenyl-4-aminophenyl)-N and N' - diphenyl -4 and 4' -- the - diamino -1 and 1' - biphenyl film (following and TPD232 film) was formed. This TPD232 film functions as a hole injection layer. Next, the above-mentioned electron hole transportability compound (A-2) of 20nm of thickness was formed on TPD232 film. This compound (A-2) film functions as an electron hole transportation layer. Furthermore, the tris (eight quinolinol) aluminum film (henceforth, Alq film) of 40nm of thickness was formed on the compound (A-2) film.

This Alq film functions as a luminous layer. next -- Li (the source of Li: SAESU getter company make), and Alq -- duality -- it was made to vapor-deposit and the Alq:Li film was formed by 20nm of thickness as an electronic injection layer (cathode). On this Alq:Li film, Metal aluminum was made to vapor-deposit, metal cathode was formed, and the organic EL device was produced.

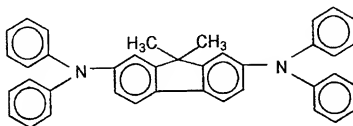
As for this component, blue luminescence of luminescence brightness 153 cd/m², the maximum luminescence brightness of 50000 cds/m², and luminous efficiency 3.2 cd/A was obtained by direct-current-voltage 6V. Moreover, it saved in the 100-degree C environment as a heat-resistant retention test for 500 hours. When direct-current-voltage 6V were impressed like trial before, 98% of brightness was shown to early brightness, and it was 98% of brightness retention.

[0035]

The example 1 of a comparison

It sets in the example 1 and is the following compound TPAF (glass transition temperature of less than 100 degrees C) instead of a compound (A-2).

[Formula 8]



After having produced the organic EL device similarly, having measured luminescence brightness and luminous efficiency, observing the luminescent color and saving under the temperature of 85 degrees C as a heat-resistant test further by direct-current-voltage 5V except for having used it for 500 hours, the brightness retention from initial brightness was measured. The result is shown in Table 1.

[0036]

[Table 1]

表 1

	化合物の種類	電圧 (V)	発光輝度 (cd/m ²)	発光効率 (cd/A)	発光色	輝度保持率 (%)
比較例 1	TPAF	5	1 5 0	2 . 5	緑	5 6

[0037]

As shown in Table 1, the organic EL device using the new arylamine compound of this invention has luminescence brightness and high luminous efficiency, and is excellent in thermal resistance. This has a glass transition temperature of the new arylamine compound of this invention as high as 100 degrees C or more, and it is for not interacting with a luminous layer.

[0038]

[Effect of the Invention]

As mentioned above, as explained to the detail, it is high brightness, and the organic electroluminescent element using the new arylamine compound of this invention has high thermal resistance, and is long lasting, and electron hole transportability is excellent, and it is high luminous efficiency.

For this reason, the organic electroluminescent element of this invention is useful as the light source of the flat-surface illuminant of wall tapestry television, the back light of a display, etc.

[Translation done.]

*** NOTICES ***

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.*** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

TECHNICAL FIELD

[Field of the Invention]

this invention -- a new arylamine compound and an organic electroluminescent element -- being related -- especially -- high brightness -- it is -- thermal resistance -- high -- long lasting -- electron hole transportability -- excelling -- high -- it is related with the new arylamine compound which realizes luminous efficiency organic electroluminescent element and it.
[0002]

[Translation done.]

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.**** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

PRIOR ART

[Description of the Prior Art]

The organic electroluminescence (EL) component which used the organic substance is used as the light source of the flat-surface illuminant of wall tapestry television, the back light of a display, etc., and development is performed briskly.

the electroluminescence phenomenon of an organic material will be observed by Pope and others (Pope) with an anthracene single crystal in 1963 (J. Chem.Phys.38 (1963) 2042) -- HERUFURIHII (Helfinck) and Shneider (Schneider) have succeeded in observation of comparatively strong injection EL by using a solution electrode system with sufficient injection efficiency in 1965 (Phys.Rev.Lett.14 (1965) 229). As reported since then, research which formed the organic luminescence matter with the conjugate organic host substance and the organic activator conjugate [with the condensation benzene ring] is done.

Naphthalene, an anthracene, a phenanthrene, tetracene, a pyrene, A benzopyrene, a chrysene, picene, a carbazole, a fluorene, a biphenyl, Terphenyl, triphenylene oxide, dihalo biphenyl, trans-stilbene and 1, and 4-diphenyl butadiene etc. was shown as an example of an organic host substance, and an anthracene, tetracene, pentacene, etc. were mentioned as an example of an activator. However, each of these organic luminescence matter existed as a monolayer with the thickness exceeding 1 micrometer, and needed high electric field for luminescence. For this reason, research of the thin film by the vacuum deposition method was advanced (for example, Thin Solid Films 94 (1982) 171). . However, although thin-film-izing was effective in reduction of driver voltage, it did not come to obtain the component of high brightness of practical use level.

Then, tongues (Tang) devised the EL element which carried out the laminating of the two very thin film (an electron hole transportation layer and luminous layer) with vacuum deposition between an anode plate and cathode, and realized high brightness by low driver voltage (Appl.Phys.Lett.51 (1987) 913 or U.S. Pat. No. 4356429 number). Then, as a result of furthering development of the organic compound used for an electron hole transportation layer and a luminous layer for about ten years, the life and luminous efficiency of utilization level were attained. Consequently, as for the organic EL device, utilization is started from the display of a car stereo and a cellular phone etc.

[0003]

However, in the practical use side, luminescence brightness, the endurance of degradation with the passage of time to long duration use, etc. are not enough, and the further improvement is called for. It is [as opposed to / when the application to a full color display etc. is considered especially / each color of R, G, and B] 300 cd/m². Reaching the reduction-by-half life of thousands of hours or more by the above high brightness is called for. Since the energy gap of a luminous layer was as large as 2.8eV or more for blue luminescence realizing this and carrying out blue luminescence especially of the difficult thing and the energy barrier in the case of the hole injection between an electron hole transportation layer and a luminous layer was large, the field strength impressed to an interface was large, in the conventional electron hole transportation layer, a hole injection was not completed in stability but amelioration was called for.

Moreover, when premised on carrying out vehicle loading of the organic EL device, it is pointed out that

a problem is in elevated-temperature shelf-life ability 100 degrees C or more. Although it is pointed out that glass transition temperature is low in the conventional electron hole transportation layer also in this case and it tended to correspond only by improving this at 100 degrees C or more, good shelf-life ability [in / it is inadequate and / an elevated temperature] was not yet realized. Furthermore, exciplex was generated as an interaction of an electron hole transportation layer and a luminous layer, and there was also a problem that the brightness of a component deteriorated.
[0004]

[Translation done.]

*** NOTICES ***

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.*** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

EFFECT OF THE INVENTION

[Effect of the Invention]

As mentioned above, as explained to the detail, it is high brightness, and the organic electroluminescent element using the new arylamine compound of this invention has high thermal resistance, and is long lasting, and electron hole transportability is excellent, and it is high luminous efficiency.

For this reason, the organic electroluminescent element of this invention is useful as the light source of the flat-surface illuminant of wall tapestry television, the back light of a display, etc.

[Translation done.]

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.**** shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention]

what was made in order that this invention might solve the aforementioned technical problem -- it is -- high brightness -- it is -- thermal resistance -- high -- long lasting -- electron hole transportability -- excelling -- high -- it aims at offering the new arylamine compound which realizes luminous efficiency organic electroluminescent element and it.

[0005]

[Translation done.]

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

MEANS

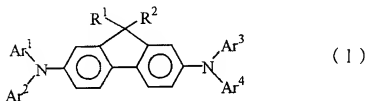
[Means for Solving the Problem]

this invention persons came to complete header this invention for the brightness of an organic electroluminescent element, thermal resistance, and a life improving, if the new arylamine compound which has specific structure is added in an organic compound layer, and electron hole transportability improving further, and becoming high luminous efficiency, as a result of repeating research wholeheartedly that the organic electroluminescent element (the following, organic EL device) which has the aforementioned desirable property should be developed.

[0006]

That is, this invention offers the new arylamine compound expressed with the following general formula (1).

[Formula 2]



[0007]

(R1 and R2 express independently the aryloxy group of the carbon atomic numbers 6-40 which are not permuted [the arylated alkyl radical of the carbon atomic numbers 7-40 which are not permuted / the aryl group of the carbon atomic numbers 6-40 which are not permuted / the alkoxy group of the carbon atomic numbers 1-30 which are not permuted / the alkyl group of the carbon atomic numbers 1-30 which are not permuted / a permutation or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or / among a formula, respectively.)

Ar1 -Ar4 Independently, the aryl group of the carbon atomic numbers 6-40 which are not permuted [a permutation or] is expressed, and even if respectively the same, you may differ, respectively. However, Ar1 -Ar4 At least two are m-biphenyl which is not permuted [a permutation or] inside, and the remainder is a biphenyl which is not permuted [a permutation or].

Moreover, this invention is an organic EL device which has an organic compound layer in inter-electrode [of a pair], and provides the charge of organic EL device material which consists of a new arylamine compound expressed with a general formula (1), and a list also with the organic EL device characterized by this organic compound layer containing said charge of organic EL device material.

[0008]

[Embodiment of the Invention]

The new arylamine compound of this invention is expressed with the above-mentioned general formula (1).

It sets to a general formula (1) and is R1. And R2 The aryloxy group of the carbon atomic numbers 6-40 which are not permuted [the arylated alkyl radical of the carbon atomic numbers 7-40 which are not permuted / the aryl group of the carbon atomic numbers 6-40 which are not permuted / the alkoxy group of the carbon atomic numbers 1-30 which are not permuted / the alkyl group of the carbon atomic numbers 1-30 which are not permuted / a permutation or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or /] is expressed independently, respectively.

As an alkyl group, for example as an alkoxy group, methyl, ethyl, n-propyl, iso-propyl, etc. As methoxy, ethoxy **, and an aryl group, as an arylated alkyl radical, phenyl, a biphenyl, naphthyl, etc. Benzyl, alpha-methylbenzyl, alpha-ethyl benzyl, alpha, and alpha-dimethylbenzyl, As an aryloxy group, 4-methylbenzyl, 4-ethyl benzyl, 2-tert-butylbenzyl, 4-n-octylbenzyl, naphthyl methyl, diphenyl methyl, etc. Phenoxy, naphthyl oxy one, and ANSUKURU oxy-** pyrenyl oxy-** biphenyl oxy-** fluoran thenyl oxy-** KURISE nil oxy-** peri RENIRU oxy-** are mentioned.

[0009]

As a substituent of each [these] radical, for example Moreover, a fluorine atom, a chlorine atom, Halogen atoms, such as a bromine atom and iodine atom, methyl, ethyl, n-propyl, Alkyl groups, such as iso-propyl, methoxy, the alkoxy group of ethoxy **, Arylated alkyl radicals, such as aryloxy groups, such as phenoxy, benzyl, phenethyl, and phenylpropyl, A nitro group, a cyano group, a dimethylamino radical, a dibenzylamino radical, a diphenylamino radical, Heterocycle radicals, such as aryl groups, such as permutation amino groups, such as a morpholino group, a phenyl group, a toluyl radical, a biphenyl radical, a naphthyl group, an anthryl radical, and a pyrenyl radical, a pyridyl radical, a thienyl group, a furil radical, a quinolyl radical, and a cull PAZORIRU radical, etc. are mentioned.

[0010]

Moreover, it sets to a general formula (1) and is Ar1 -Ar4. Independently, the aryl group of the carbon atomic numbers 6-40 which are not permuted [a permutation or] is expressed, and even if respectively the same, you may differ, respectively.

As an aryl group, a phenyl group, a toluyl radical, a biphenyl radical, a naphthyl group, an anthryl radical, a pyrenyl radical, etc. are mentioned, for example.

It sets to a general formula (1) and is Ar1 -Ar4. At least two are m-biphenyl which is not permuted [a permutation or] inside, and the remainder is a biphenyl which is not permuted [a permutation or].

[0011]

Ar1 -Ar4 As a substituent in the radical to express For example, halogen atoms, such as a fluorine atom, a chlorine atom, a bromine atom, and iodine atom, Alkyl groups, such as methyl, ethyl, n-propyl, and iso-propyl, Aryloxy groups, such as methoxy, an alkoxy group of ethoxy **, and phenoxy, Arylated alkyl radicals, such as benzyl, phenethyl, and phenylpropyl, A nitro group, a cyano group, a dimethylamino radical, a dibenzylamino radical, a diphenylamino radical, Heterocycle radicals, such as aryl groups, such as permutation amino groups, such as a morpholino group, a phenyl group, a toluyl radical, a biphenyl radical, a naphthyl group, an anthryl radical, a pyrenyl radical, and a fluoran thenyl radical, a pyridyl radical, a thienyl group, a furil radical, a quinolyl radical, and a cull PAZORIRU radical, etc. are mentioned.

Furthermore, as an aryl substituent in an aryl permutation biphenyl, a phenyl group, a biphenyl radical, a terphenyl radical, a naphthyl group, an anthryl radical, a fluorenyl group, etc. are mentioned, for example.

Ar1 And Ar3 m-biphenyl which is not permuted [a permutation or] and Ar2 And Ar4 It is desirable that it is the biphenyl which is not permuted [a permutation or].

[0012]

The organic EL device of this invention is an organic EL device which has an organic compound layer in inter-electrode [of a pair], and this organic compound layer contains the charge of organic EL device material of said new arylamine compound.

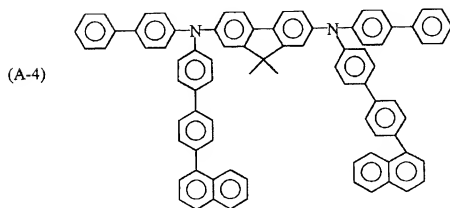
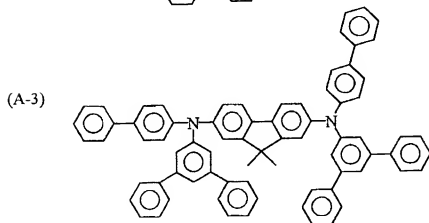
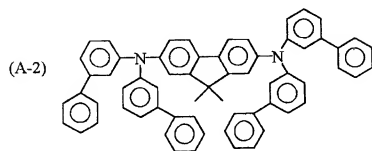
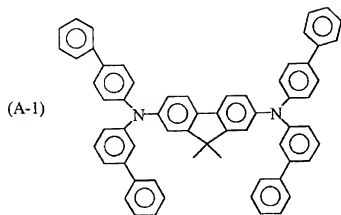
It is desirable that said organic compound layer is a luminous layer or an electron hole transportation layer. Moreover, it is also desirable to have the layer in which said organic compound layer contains the charge of organic EL device material and luminescent material of said new arylamine compound. it is because at least non-***** which it has a high glass transition point that the brightness of an organic EL device, thermal resistance, a life, and luminous efficiency improve, and it cannot interact with luminescent material easily when this arylamine compound is excellent in electron hole transportability and a hole injection is possible for stability, and is produced by the interaction will be avoided if an organic compound layer boils said new arylamine compound further at least and it is made to contain.

[0013]

Although the example of representation of a general formula (1) is illustrated in the new arylamine compound of this invention below, this invention is not limited to these examples of representation.

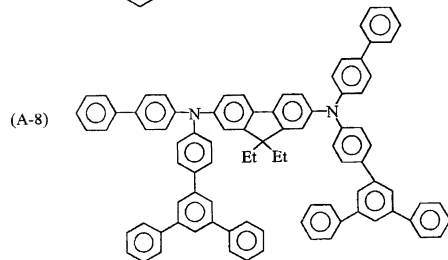
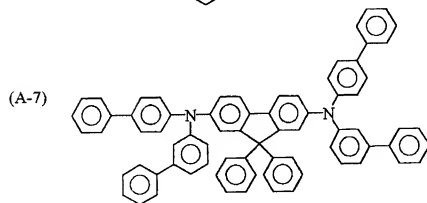
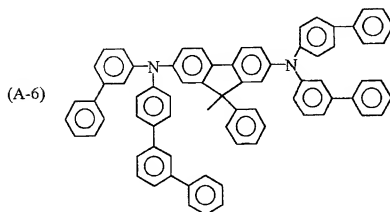
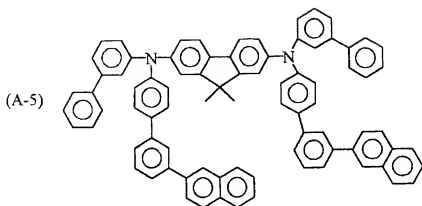
[0014]

[Formula 3]



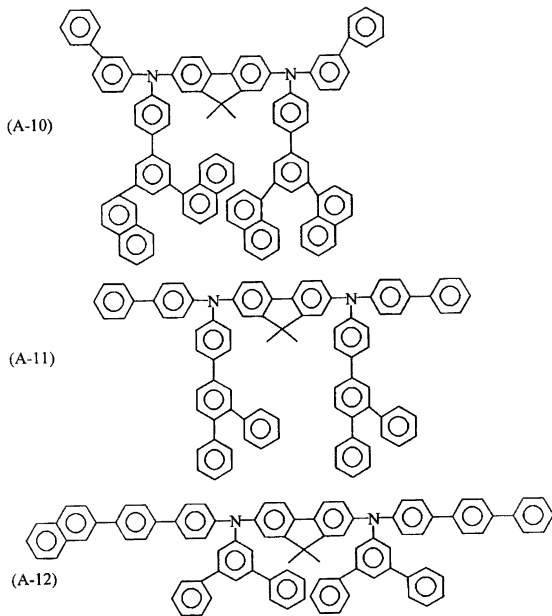
[0015]

[Formula 4]



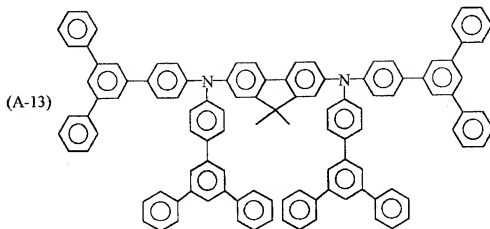
[0016]

[Formula 5]



[0017]

[Formula 6]



[0018]

The organic EL device of this invention is a component in which the monolayer or the multilayer organic compound layer was formed between an anode plate and cathode. In the case of the monolayer mold, the luminous layer is prepared between an anode plate and cathode. A luminous layer contains luminescent material, and in order to make the electron hole which was poured in from the anode plate in addition to it, or the electron poured in from cathode convey to luminescent material, it may contain a hole-injection ingredient or an electron injection ingredient. However, as for luminescent material, it is desirable to have very high fluorescence quantum efficiency, high electron hole transport capacity, and electronic transport capacity, and to form a uniform thin film. The organic EL device of a multilayer mold has some which carried out the laminating with the multilayer configuration of (an anode plate / hole injection layer / luminous layer / cathode), (an anode plate / luminous layer / electronic injection layer / cathode), and (an anode plate / hole injection layer / luminous layer / electronic injection layer / cathode).

[0019]

In addition to the new arylamine compound of this invention, the further well-known luminescent material, a doping ingredient, a hole-injection ingredient, and an electron injection ingredient can also be used for a luminous layer if needed. As a desirable usage of this new arylamine compound, it adds at 0.5 - 100 % of the weight of concentration in the layer of either a luminous layer, an electronic injection layer, an electron hole transportation layer or a hole injection layer. It is 50 - 100 % of the weight of concentration still more preferably.

An organic EL device can prevent the fall of the brightness by quenching, or a life by making it multilayer structure. If there is need, it can be used combining luminescent material, other doping ingredients, a hole-injection ingredient, or an electron injection ingredient. Moreover, improvement in luminescence brightness or luminous efficiency and luminescence of red or white can also be obtained with other doping ingredients. Moreover, a hole injection layer, a luminous layer, and an electronic injection layer may be formed of the lamination more than a bilayer, respectively. In the case of a hole injection layer, in that case, the layer which conveys [the layer which pours in an electron hole from an electrode] an electron hole for an electron hole from a hole injection layer and a hole injection layer to a reception luminous layer is called an electron hole transportation layer. Similarly, in the case of an electronic injection layer, the layer which conveys [the layer which pours in an electron from an electrode] an electron for an electron from an electronic injection layer and an electronic injection layer to a reception luminous layer is called an electron transport layer. These each class is used by each factor, such as adhesion with the energy level of an ingredient, thermal resistance, an organic compound

layer, or a metal electrode, choosing it.

[0020]

As the luminescent material which can be used for an organic compound layer with said new arylamine compound, or a host ingredient, there is condensed multi-ring aromatic series, for example, an anthracene, naphthalene, a phenanthrene, a pyrene, tetracene, pentacene, coronene, a chrysene, a fluorescein, perylene, rubrene, and those derivatives exist. Non [furthermore, / phthaloperylene, naphthaloperylene, and peri non, / phthaloperi] Naphthaloperi non, a diphenyl butadiene, a tetraphenyl butadiene, A coumarin, oxadiazole, aldzine, bis-benzoKISAZORIN, Bis-styryl, pyrazine, a cyclopentadiene, a quinoline metal complex, An amino quinoline metal complex, a benzoquinoline metal complex, an imine, diphenylethylene, Although a vinyl anthracene, a diamino carbazole, a pyran, thiopyran, poly methine, merocyanine, an imidazole chelation oxy-NOIDO compound, Quinacridone, rubrene, a stilbene system derivative, a fluorochrome, etc. are mentioned, it is not limited to these.

[0021]

The compound which has the capacity to convey an electron hole, as a well-known hole-injection ingredient, has the hole-injection effectiveness which was excellent to the hole-injection effectiveness, the luminous layer, or luminescent material from an anode plate, and prevented migration into the electronic injection layer or electron injection ingredient of an exciton generated by the luminous layer, and was excellent in the thin film organization potency force is desirable. Specifically A phthalocyanine derivative, a naphthalocyanine derivative, a porphyrin derivative, Oxazole, oxadiazole, triazole, an imidazole, imidazolone, Imidazole thione, pyrazoline, a pyrazolone, a tetrahydro imidazole, Oxazole, oxadiazole, a hydrazone, an acyl hydrazone, The poly aryl alkane, a stilbene, a butadiene, a benzidine mold triphenylamine, a styryl amine mold triphenylamine, a diamine mold triphenylamine, etc., Although polymeric materials, such as those derivatives and a polyvinyl carbazole, polysilane, and a conductive polymer, are mentioned, it is not limited to these.

[0022]

The still more effective hole-injection ingredient in the well-known hole-injection ingredient which can be used in the organic EL device of this invention is the third class amine derivative of aromatic series, or a phthalocyanine derivative.

The example of the third class amine derivative of aromatic series A triphenylamine, a tritolyl amine, A tolyl diphenylamine, N, N'-diphenyl-N, N' - (3-methylphenyl) -1, the 1'-biphenyl -4, 4'-diamine, N, N, N', N' - (4-methylphenyl) -1, the 1'-phenyl -4, 4'-diamine, N, N, N', N' - (4-methylphenyl) -1, the 1'-biphenyl -4, 4'-diamine, N and N' - diphenyl-N and N' - dinaphthyl -1 and 1' -- the - biphenyl -4 and 4' - diamine -- N, N'-(methylphenyl)-N, N' -(4-n-butylphenyl)- A phenanthrene -9, 10-diamine, Although it is oligomer or a polymer with the third class amines frame of such aromatic series, such as an N and N-bis(4-G 4-tolylamino phenyl)-4-phenyl-cyclohexane, it is not limited to these.

As for the example of a phthalocyanine (Pc) derivative, ***** is not limited to these with a phthalocyanine derivative and naphthalocyanine derivatives, such as H2 Pc, CuPc, CoPc, NiPc, ZnPc, PdPc, FePc, MnPc, ClAlPc, ClGaPc, ClInPc, ClSnPc, Cl2 SiPc, (HO) AlPc, (HO) GaPc, VOPc, TiOPc, MoOPc, and GaPc-O-GaPc.

[0023]

The compound which has the capacity to convey an electron, as a well-known electron injection ingredient, has the electron injection effectiveness which was excellent to the electron injection effectiveness, the luminous layer, or luminescent material from cathode, and prevented migration to the hole injection layer of the exciton generated by the luminous layer, and was excellent in the thin film organization potency force is desirable. concrete -- full -- me -- non, although anthra quinodimethan, diphenquinone, thiopyran dioxide, oxazole, oxadiazole, triazole, an imidazole, perylene tetracarboxylic acid, deflection ORENIRIDEN methane, anthra quinodimethan, anthrones, etc. and those derivatives are mentioned, it is not limited to these. Moreover, charge impregnation nature can also be raised by adding the electronic acceptance matter into a hole-injection ingredient, and adding the electron-donate matter into an electron injection ingredient.

[0024]

In the organic EL device of this invention, a still more effective well-known electron injection ingredient is a metal complex compound or a nitrogen-containing five membered ring derivative.

The example of a metal complex compound 8-hydroxyquinolate lithium, Bis(8-hydroxyquinolate) zinc, bis(8-hydroxyquinolate)copper, Bis(8-hydroxyquinolate) manganese, tris(8-hydroxyquinolate)aluminum, Tris (2-methyl-8-hydroxyquinolate) aluminum, A tris (8-hydroxyquinolate) gallium, bis(10-hydroxy benzo[h] quinolate) beryllium, Bis(10-hydroxy benzo[h] quinolate) zinc, a bis(2-methyl-8-quinolate) chloro gallium, Although a bis(2-methyl-8-quinolate) (o-cresolate) gallium, bis(2-methyl-8-quinolate) (1-naphth RATO) aluminum, a bis(2-methyl-8-quinolate) (2-naphth RATO) gallium, etc. are mentioned It is not limited to these.

[0025]

Moreover, a nitrogen-containing 5 member derivative has oxazole, a thiazole, oxadiazole, thiadiazole, or a desirable triazole derivative. Specifically, it is 2 and 5-screw (1-phenyl). - 1, 3, 4-oxazole, Dimethyl POPOP, 2, 5-screw (1-phenyl) - 1, 3, 4-thiazole, 2, 5-screw (1-phenyl) - 1, 3, 4-oxadiazole, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl) 1, 3, 4-oxadiazole, 2, 5-bis(1-naphthyl)-bis[1, 3, 4-oxadiazole, 1, and 4-] [2- (5-phenyl oxadiazolyl)] benzene, 1, 4-screw [2- (5-phenyl oxadiazolyl)-4-tert-butylbenzene], 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)- 1, 3, and 4-thiadiazole -- 2, 5-bis(1-naphthyl)-bis[1, 3, 4-thiadiazole, 1, and 4-] [2- (5-phenyl thiadiazolyl)] benzene, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-, although 1, 3, 4-triazole, 2, and 5-bis(1-naphthyl)-bis[1, 3, 4-triazole, 1, and 4-] [2- (5-phenyl triazolyl)] benzene etc. is mentioned It is not limited to these.

In this invention, an inorganic compound layer may be prepared between a luminous layer and an electrode for the improvement in charge injectional. as such an inorganic compound layer -- alkali metal compounds (a fluoride, oxide, etc.), an alkaline-earth-metal compound, etc. -- it is -- concrete -- LiF, Li2O, RaO, SrO and BaF2, and SrF2 etc. -- it is mentioned.

[0026]

What has a bigger work function than 4eV as a conductive ingredient used for the anode plate of an organic EL device is suitable, and organic conductive resin, such as the poly thiophene and polypyrrole, is used for gold oxide groups, such as tin oxide used for those alloys, such as carbon, aluminum, vanadium, iron, cobalt, nickel, a tungsten, silver, gold, platinum, and palladium, and an ITO substrate, and a NESA substrate, and indium oxide, and a pan. Although what has a work function smaller than 4eV as conductive matter used for cathode is suitable and those alloys, such as magnesium, calcium, tin, lead, titanium, an yttrium, a lithium, a ruthenium, manganese, and aluminum, are used, it is not limited to these. As an alloy, although magnesium/silver, magnesium/indium, a lithium/aluminum, etc. are mentioned as an example of representation, it is not limited to these. The ratio of an alloy is controlled by the temperature of the source of vacuum evaporation, the ambient atmosphere, a degree of vacuum, etc., and is chosen as a suitable ratio. As long as an anode plate and cathode have the need, they may be formed of the lamination more than a bilayer.

[0027]

In order to make light emit efficiently in an organic EL device, as for one [at least] field, it is desirable to make it transparency enough in the luminescence wavelength field of a component. Moreover, the transparent thing of a substrate is desirable. The above-mentioned conductive ingredient is used for a transparent electrode, and it sets it up so that predetermined translucency may secure by approaches, such as vacuum evaporation and sputtering. As for the electrode of a luminescence side, it is desirable to make light transmittance 10% or more. Although a substrate is not limited if it has mechanical and thermal reinforcement and has transparency, it has a glass substrate and a transparency resin film. As a transparency resin film, polyethylene, an ethylene-vinylacetate copolymer, An ethylene-vinylalcohol copolymer, polypropylene, polystyrene, Polymethylmethacrylate, a polyvinyl chloride, polyvinyl alcohol, A polyvinyl butyral, nylon, a polyether ether ketone, the poly ape phone, A polyether ape phon, a tetrafluoroethylene-perfluoroalkyl vinyl ether copolymer, Polyvinyl fluoride, a tetrafluoroethylene-ethylene copolymer, A tetrafluoroethylene-hexafluoropropylene copolymer, polychlorotrifluoroethylene resin, poly vinylidene fluoride, polyester, a polycarbonate, polyurethane, polyimide, polyether imide, polyimide, polypropylene, etc. are mentioned.

[0028]

A protective layer is prepared on the surface of a component for the improvement of stability to temperature, humidity, an ambient atmosphere, etc., or the organic EL device of this invention can also protect the whole component with a silicone oil, resin, etc.

Formation of each class of an organic EL device can apply which approach of the wet forming-membranes methods, such as the dry type forming-membranes methods, such as vacuum deposition, sputtering, plasma, and ion plating, spin coating, dipping, and flow coating. Although especially thickness is not limited, it is necessary to set it as suitable thickness. If thickness is too thick, in order to obtain a fixed optical output, big applied voltage will be needed and effectiveness will worsen. If thickness is too thin, even if a pinhole etc. will occur and it will impress electric field, sufficient luminescence brightness is not obtained. The usual thickness has the still more desirable range of 0.2 micrometers from 10nm, although the range of 10 micrometers is suitable from 5nm.

[0029]

The solvent may be any, although suitable solvents, such as ethanol, chloroform, a tetrahydrofuran, and dioxane, are made to dissolve or distribute the ingredient which forms each class in the case of the wet forming-membranes method and a thin film is formed. Moreover, also in which organic thin film layer, suitable resin and a suitable additive may be used on a membrane formation disposition for pinhole prevention of the film etc. As possible resin of use, conductive resin, such as photoconductivity resin, such as insulating resin, such as polystyrene, a polycarbonate, polyarylate, polyester, a polyamide, polyurethane, polysulfone, polymethylmethacrylate, polymethyl acrylate, and a cellulose, and those copolymers, poly-N-vinylcarbazole, and polysilane, the poly thiophene, and polypyrrole, can be mentioned. Moreover, an antioxidant, an ultraviolet ray absorbent, a plasticizer, etc. can be mentioned as an additive.

[0030]

The organic EL device of this invention can be used for the light source of the back light of flat-surface illuminants, such as a flat-panel display of a flat TV, a copying machine, a printer, and a liquid crystal display, or instruments, the plotting board, a beacon light, etc.

[0031]

[Translation done.]

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

EXAMPLE

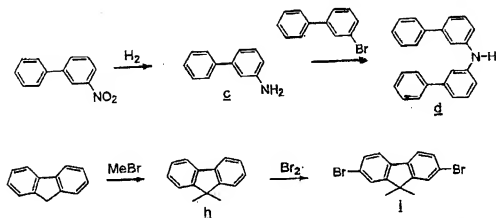
[Example]

Hereafter, this invention is further explained to a detail based on a synthetic example and an example.

The synthetic example 1 (compound (A-2))

The reaction path of intermediate field d, intermediate field h, and intermediate field i is shown below.

[Formula 7]



[0032]

Composition of intermediate field c

It was made to react for 7 hours, teaching $Pd/C(7.5\%)$ 1g and blowing hydrogen below 30 degrees C into the 3-nitro diphenyl 13g (65mmol) 75ml suspension of ethanol. Reaction mixture was filtered, and after carrying out Pd/C removal, intermediate-field c 10.8g which carries out reduced pressure distilling off of the solvent, and is made into the purpose was obtained (98% of yield).

Composition of intermediate field d

They are intermediate-product c 6.8g (40mmol), 3-BUROMO phenyl 9.2g (40mmol), and tris (JIBEN zylidene acetone) the bottom of an argon air current, and in a 300ml three necked flask with a cooling pipe. After adding JIPARAJIUMU 1.1g (1.5-mol%), tree o-toluyil phosphine 0.72g (three-mol%), t-butoxy sodium 3.8g (40mmol), and 100ml of desiccation toluene, heating **** was carried out at 100 degrees C overnight. The depositing crystal was separated after reaction termination, 100ml of methanols washed, and intermediate-field d 11.8g (90% of yield) was obtained.

[0033]

Composition of intermediate field h

Fluorene 22g (0.13mmol) and 100ml of desiccation THF were taught the bottom of an argon air current,

and into the 500ml three necked flask, and it cooled at -78 degrees C. 120ml (2.6M hexane) (0.32 mols) of n-butyl lithium was dropped here. After agitating in this ** for 1 hour, the 60ml solution of methyl bromide 28g (0.3 mols)/THF(s) was dropped at -78 degrees C. Then, it returned to the room temperature gradually and ****(ed) at the room temperature overnight. Reaction mixture was poured into 1l. of water after reaction termination, and desiccation distilling off was carried out with sulfuric anhydride magnesium after an IPE extract and saturation brine washing. The column chromatography (silica gel, an expansion solvent: hexane) refined the residue, and intermediate-field h25g (98% of yield) was obtained.

Composition of intermediate field i

Into the 1l. three necked flask which shaded, intermediate-field h9.7g (50mmol), 100ml of chloroform, and FeCl₂ 0.2g were taught. Subsequently, 24g (0.15 mols) of bromines was dropped at 0 degree C. Then, it was made to react at a room temperature overnight. Intermediate-field i15g which carries out stoving of the precipitated crystal after washing separation, rinsing, and ethanol, and targets it was obtained after reaction termination (85% of yield).

Composition of a compound (A-2)

They are intermediate-product i3.5g (10mmol), intermediate-product d6.4g (20mmol), and tris (JIBEN zylidene acetone) the bottom of an argon air current, and in a 300ml three necked flask with a cooling pipe. After adding JIPARAJIUMU0.27g (1.5-mol%), tree o-tolulyl phosphine 0.18g (three-mol%), t-butoxy sodium 1.9g (20mmol), and 100ml of desiccation toluene, heating **** was carried out at 100 degrees C overnight. The depositing crystal was separated after reaction termination, 100ml of methanols washed, and 6.6g of yellow powder was obtained. This thing was identified the compound (A-2) by measurement of NMR, IR, and FD-MS (field desorption mass spectrum) (80% of yield).

[0034]

Example 1

After performing ultrasonic cleaning for the glass substrate with an ITO transparent electrode of 25mmx75mmx1.1mm thickness (JIOMA tick company make) for 5 minutes in isopropyl alcohol, UV ozone washing was performed for 30 minutes. the field top of the side in which the substrate electrode holder of a vacuum evaporation system is equipped with the glass substrate with transparent electrode Rhine after washing, and transparent electrode Rhine is formed first -- said transparent electrode -- a wrap -- like -- carrying out -- N of 60nm of thickness, and N' - bis(N and N'-diphenyl-4-aminophenyl)-N and N' - diphenyl -4 and 4' -- the - diamino -1 and 1' - biphenyl film (following and TPD232 film) was formed. This TPD232 film functions as a hole injection layer. Next, the above-mentioned electron hole transportability compound (A-2) of 20nm of thickness was formed on TPD232 film. This compound (A-2) film functions as an electron hole transportation layer. Furthermore, the tris (eight quinolinol) aluminum film (henceforth, Alq film) of 40nm of thickness was formed on the compound (A-2) film. This Alq film functions as a luminous layer. next -- Li (the source of Li: SAESU getter company make), and Alq -- duality -- it was made to vapor-deposit and the Alq:Li film was formed by 20nm of thickness as an electronic injection layer (cathode). On this Alq:Li film, Metal aluminum was made to vapor-deposit, metal cathode was formed, and the organic EL device was produced.

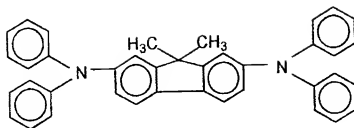
As for this component, blue luminescence of luminescence brightness 153 cd/m², the maximum luminescence brightness of 50000 cds/m², and luminous efficiency 3.2 cd/A was obtained by direct-current-voltage 6V. Moreover, it saved in the 100-degree C environment as a heat-resistant retention test for 500 hours. When direct-current-voltage 6V were impressed like trial before, 98% of brightness was shown to early brightness, and it was 98% of brightness retention.

[0035]

The example 1 of a comparison

It sets in the example 1 and is the following compound TPAF (glass transition temperature of less than 100 degrees C) instead of a compound (A-2).

[Formula 8]



After having produced the organic EL device similarly, having measured luminescence brightness and luminous efficiency, observing the luminescent color and saving under the temperature of 85 degrees C as a heat-resistant test further by direct-current-voltage 5V except for having used it for 500 hours, the brightness retention from initial brightness was measured. The result is shown in Table 1.

[0036]

[Table 1]

表 1

	化合物の種類	電圧 (V)	発光輝度 (cd/m ²)	発光効率 (cd/A)	発光色	輝度保持率 (%)
比較例 1	TPAF	5	1 5 0	2 . 5	緑	5 6

[0037]

As shown in Table 1, the organic EL device using the new arylamine compound of this invention has luminescence brightness and high luminous efficiency, and is excellent in thermal resistance. This has a glass transition temperature of the new arylamine compound of this invention as high as 100 degrees C or more, and it is for not interacting with a luminous layer.

[0038]

[Translation done.]